

Microscopie et Spectroscopie Tunnel à Balayage dans l'étude des systèmes à fortes corrélations électroniques

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François Debontridder,

Yves Noat



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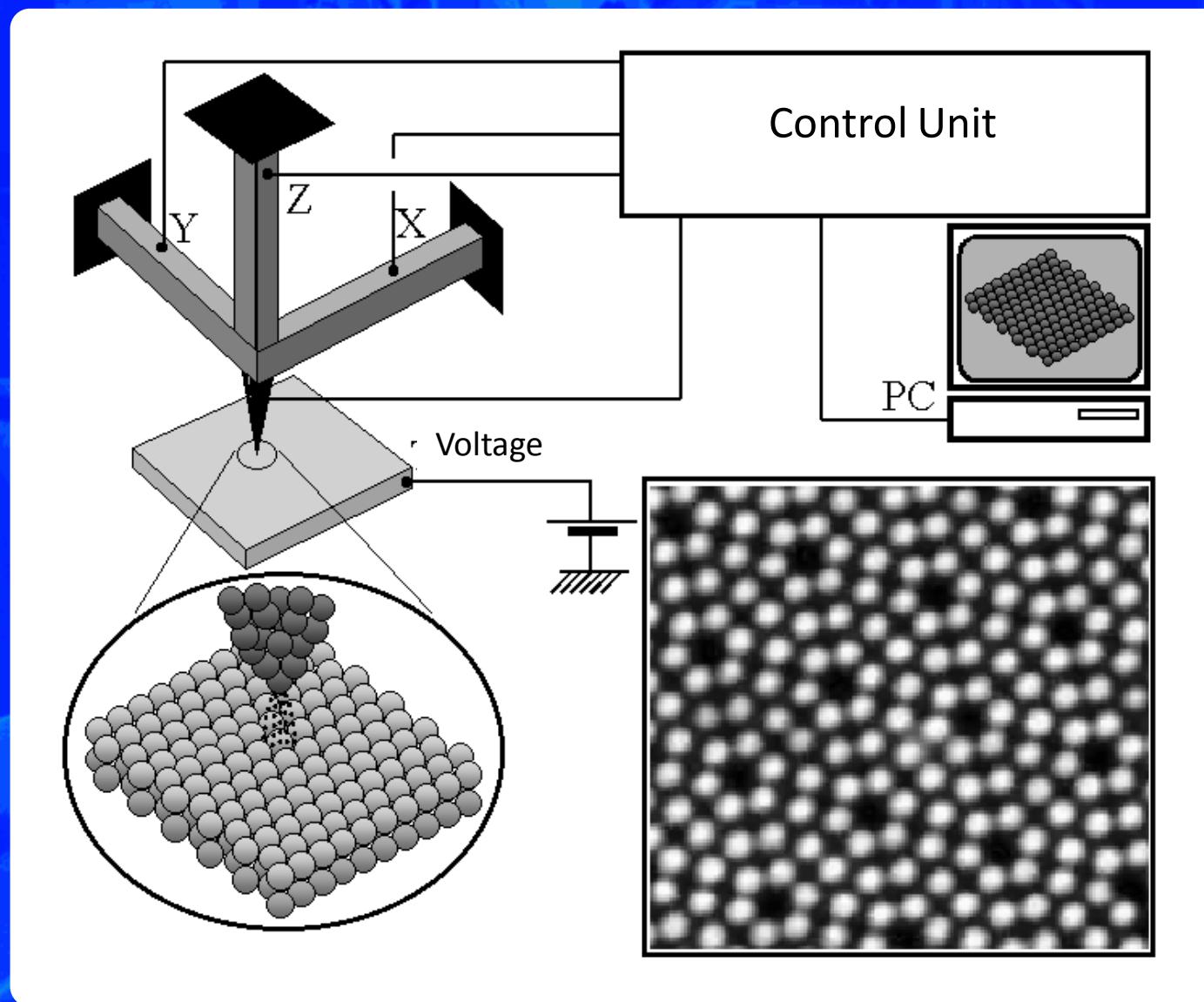
OUTLINE

Scanning Tunneling Microscopy (STM) and Spectroscopy (STS)

1. Physical background
2. How to make it, how it looks like
3. Spectroscopy of Superconductors
4. Other strongly correlated materials

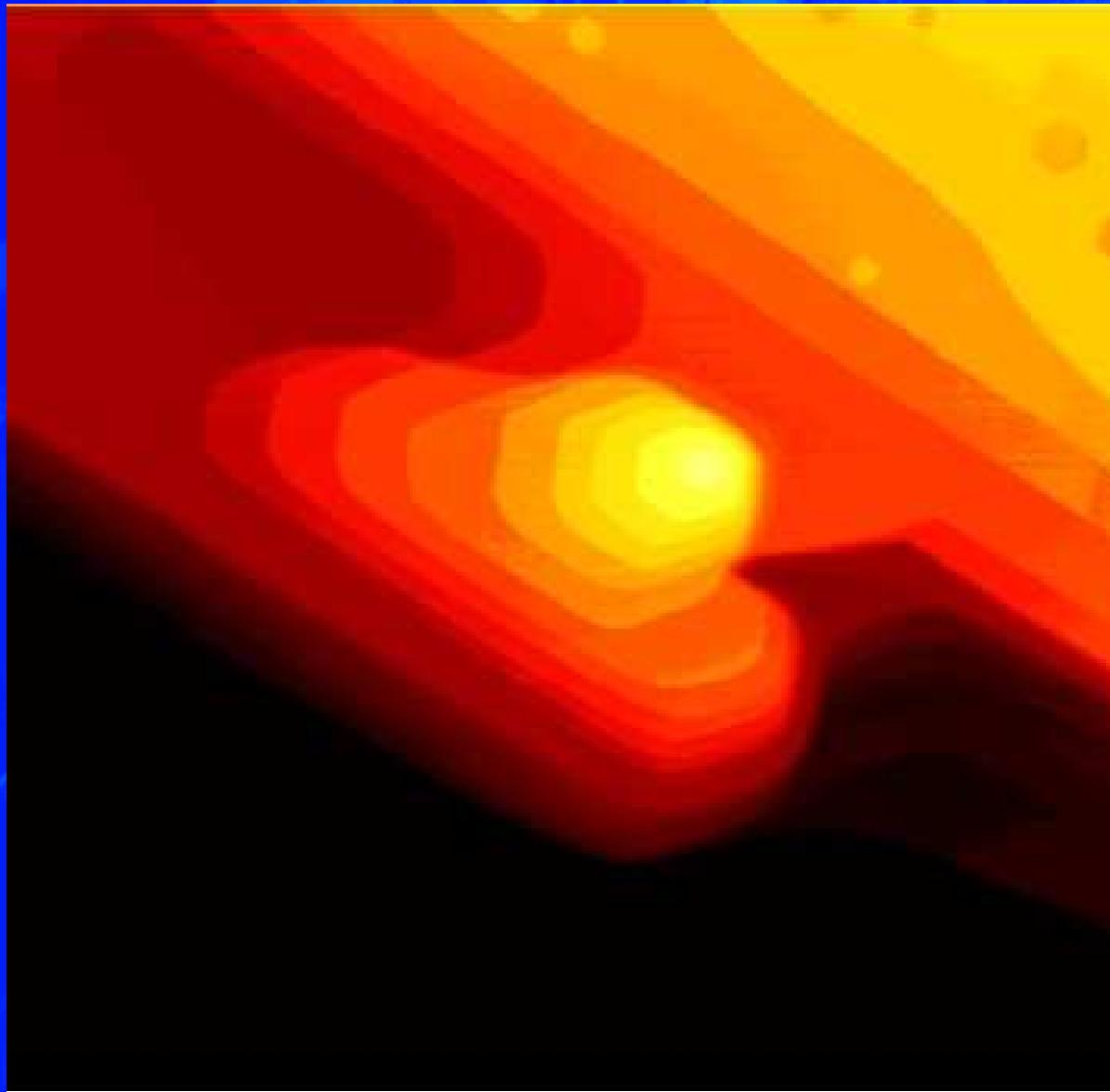
Invention du STM

*Depuis 1982 : Scanning Tunneling Microscope
par G. Binnig et H. Rohrer , Prix Nobel 1986*

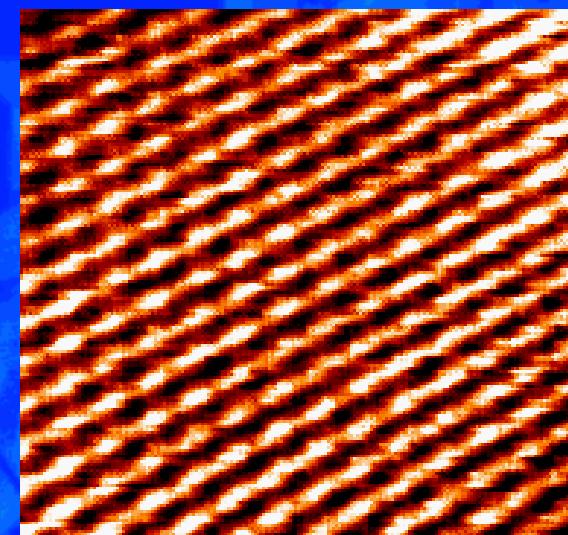


Surface Study by STM

Ag(111) at 300K



*Ag(111) at 300K:
Atomic corrugation 2pm*



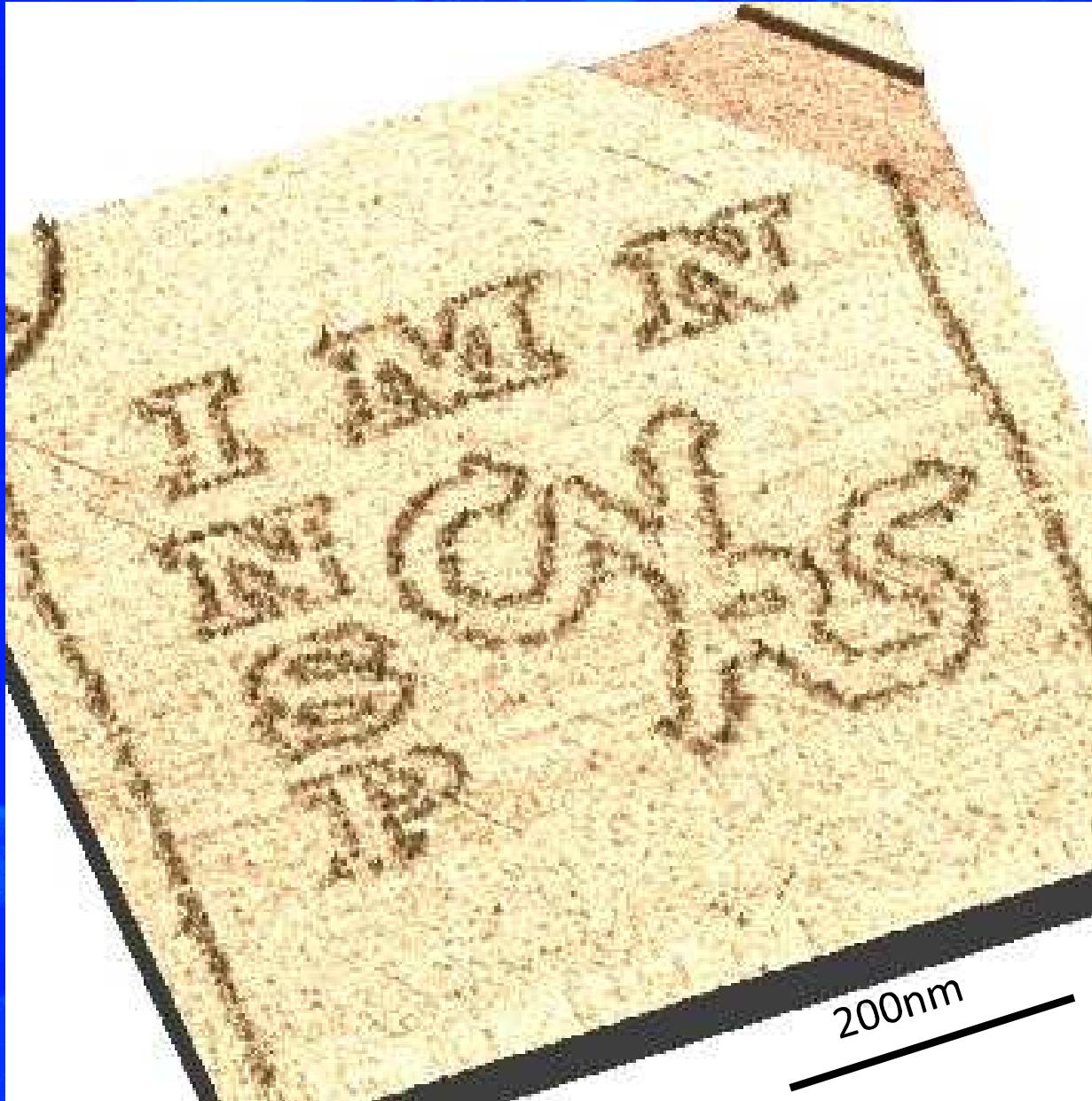
STM Controlled Growth

Pb-Morphology: 3 ML + Islands



Selection of Heights due to Z
- Quantum Confinement

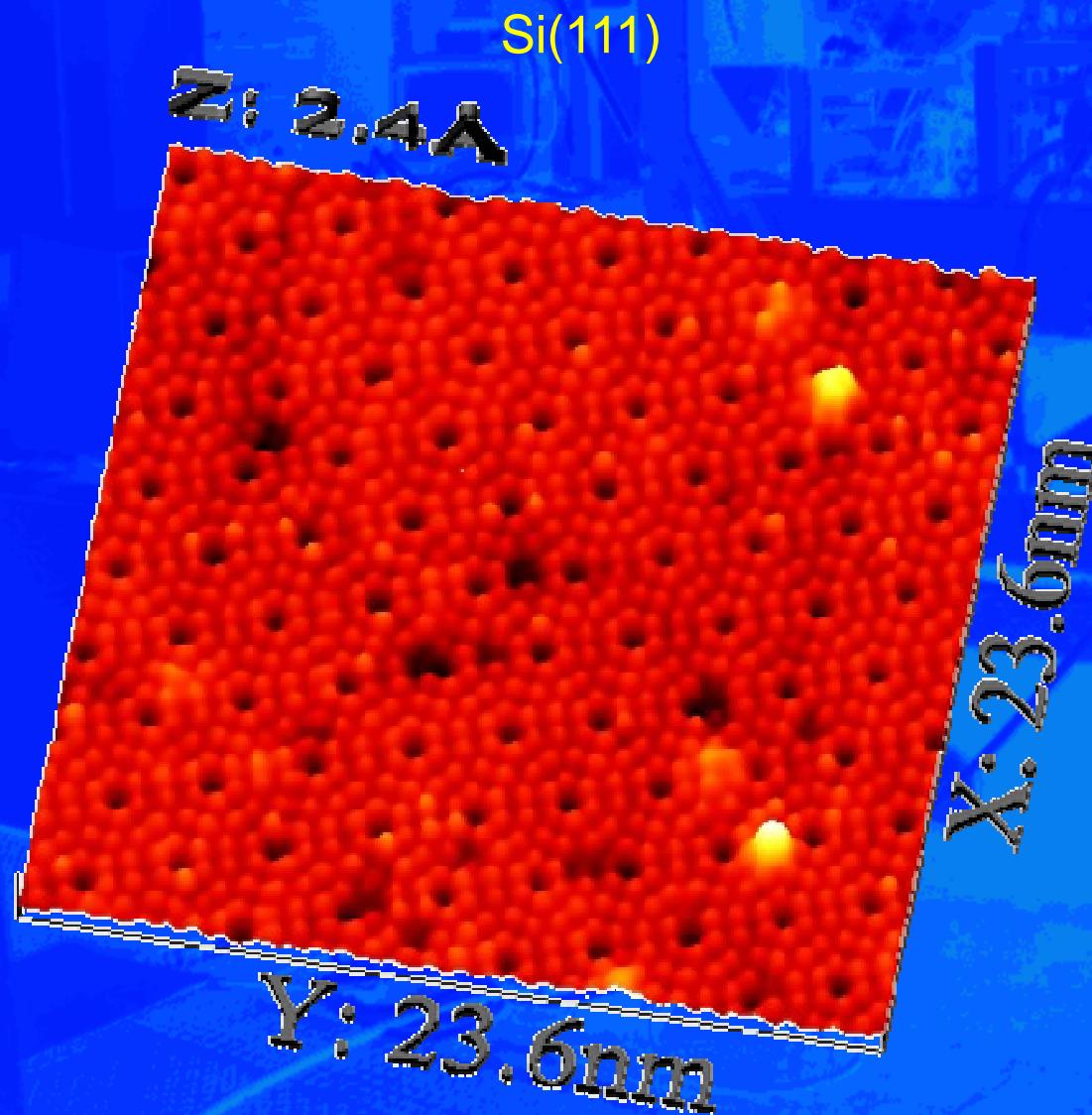
Nano-Writing with STM



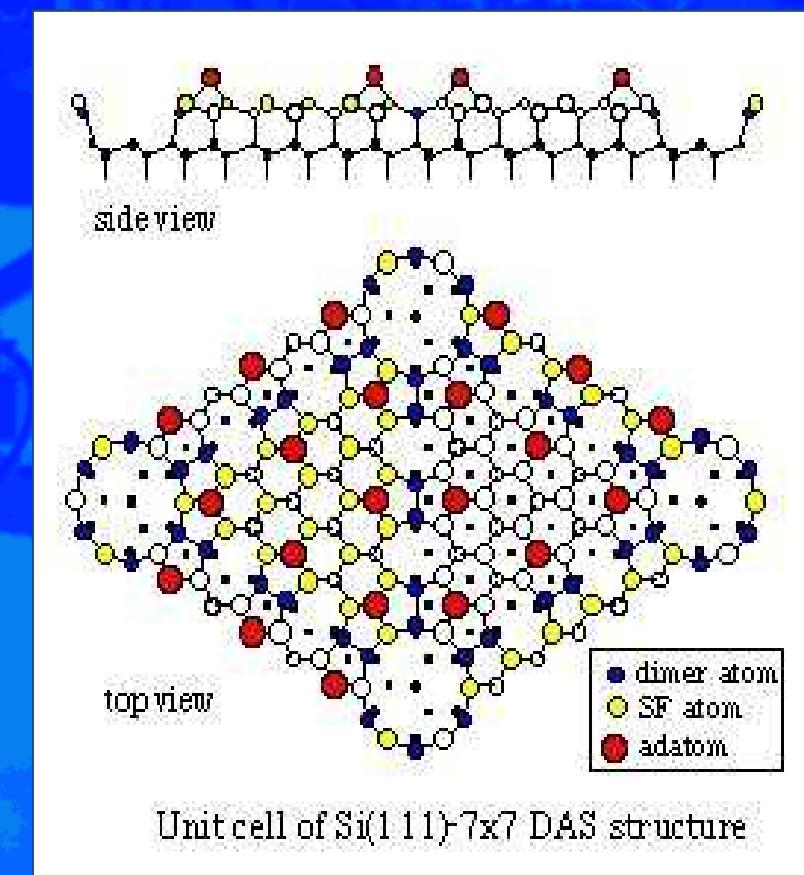
Collaboration:
L. Cario et al.
*IMN, UMR6502,
Université de Nantes*

Atomic pattern resolved by STM

Silicon: an ideal substrat

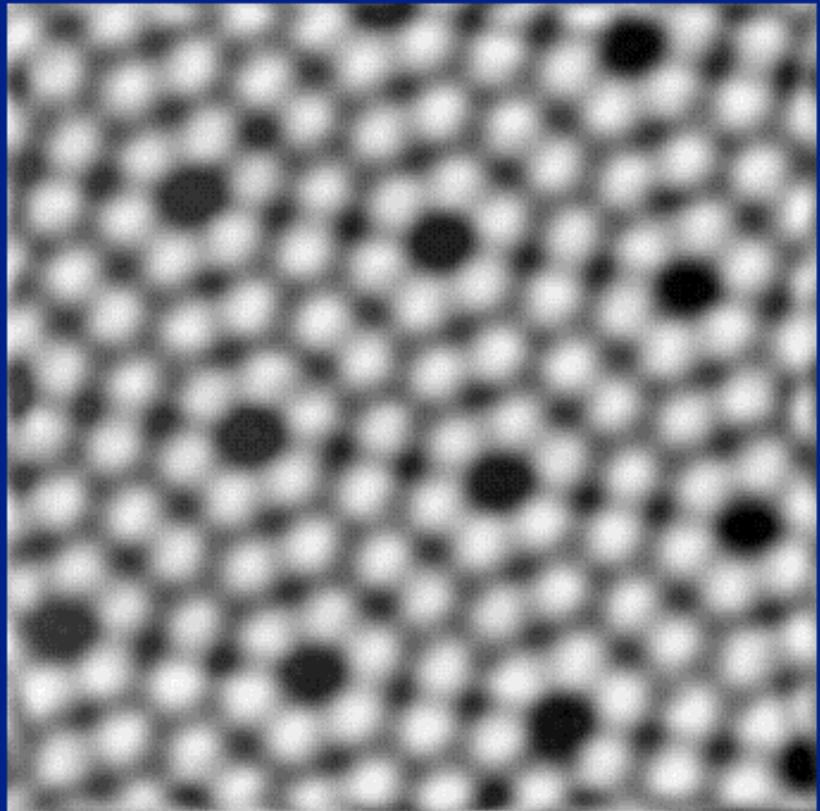


Surface Reconstruction 7x7



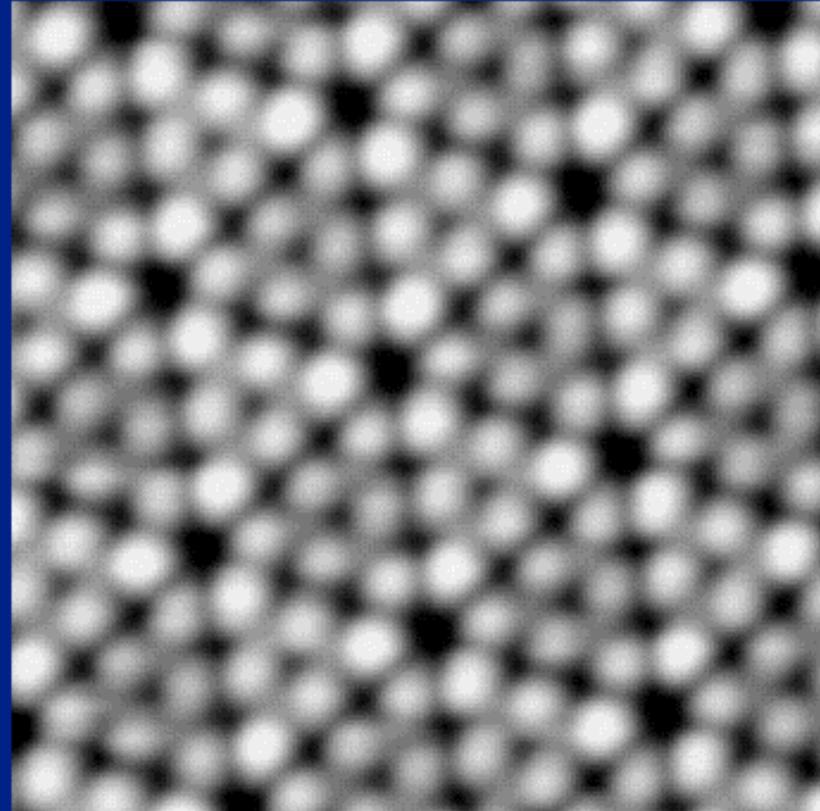
Au-delà de la topographie STM

- Si(111)-7x7 Surface



Empty-State Image

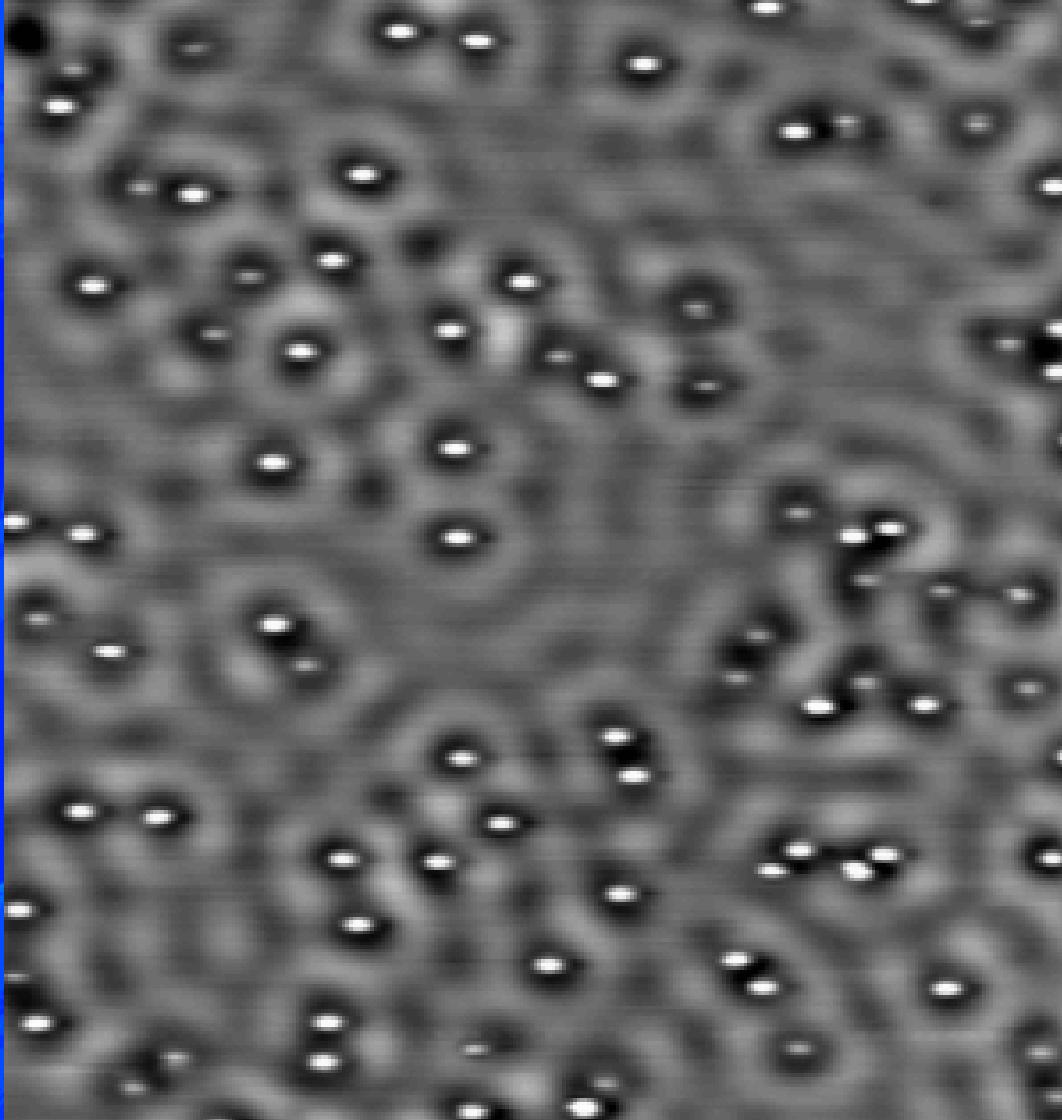
Imaging: $V_s=2V$, $I=0.6nA$



Filled-State Image

Imaging: $V_s=-2V$, $I=0.6nA$

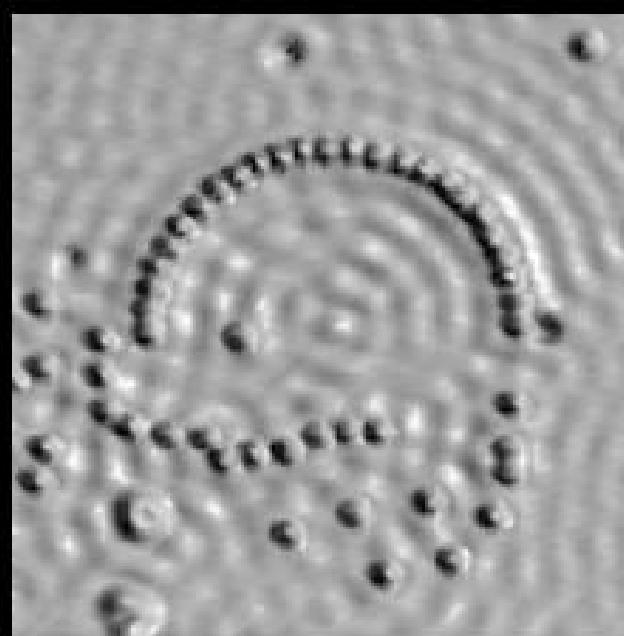
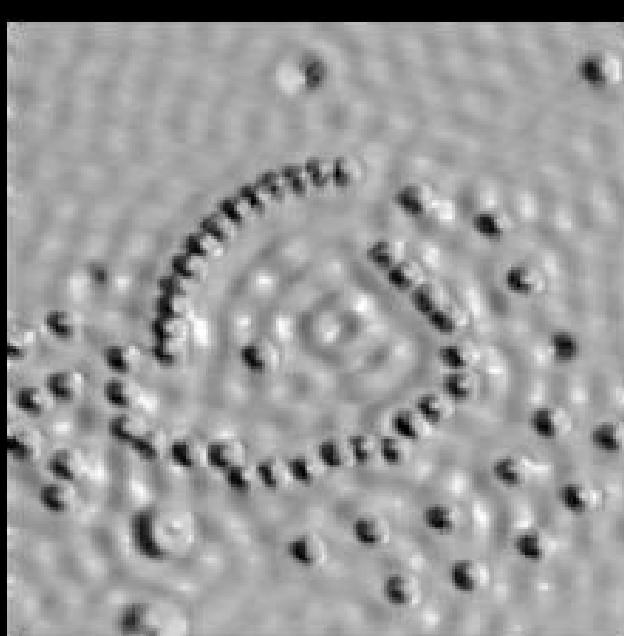
Atomes de Co sur la surface d'Ag



T. Cren et al.
EPFL Lausanne

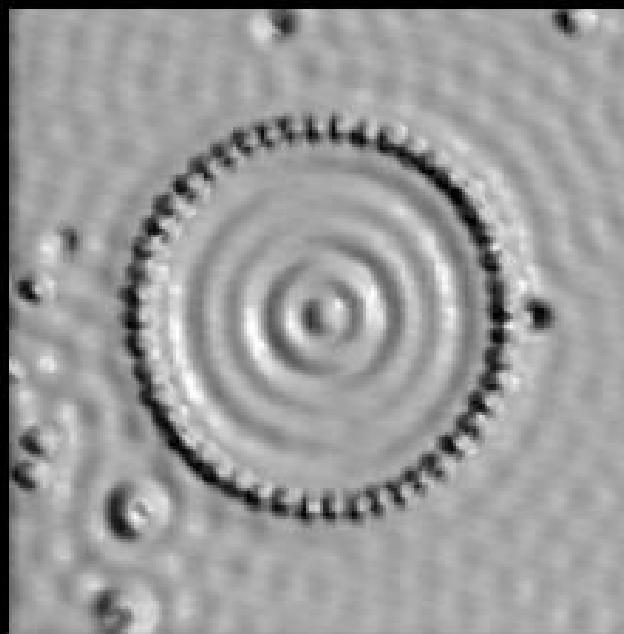
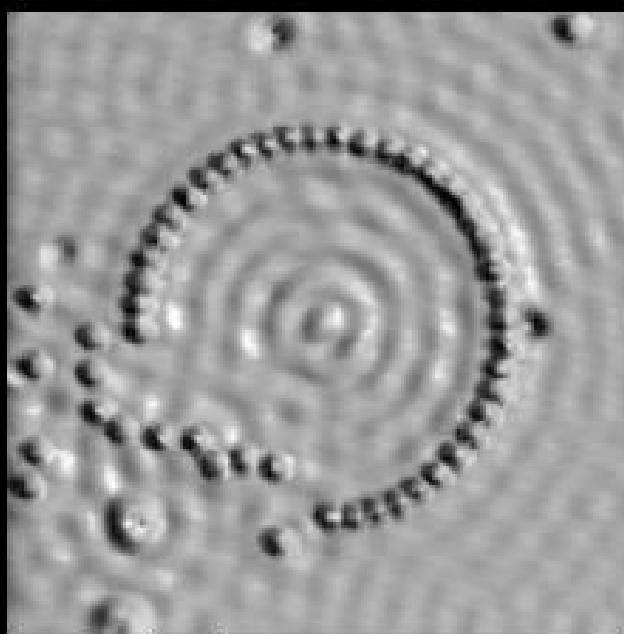
T = 30 K (-243°C)
Ultra-vide

Quantum Resonator



● Fe atoms at
Cu (111)

T = 4,2 K (-269°C)
UHV



D. Eigler et col.
*IBM Research Division,
Almaden Research
Center, California, USA*

site WEB:
<http://www.almaden.ibm.com>

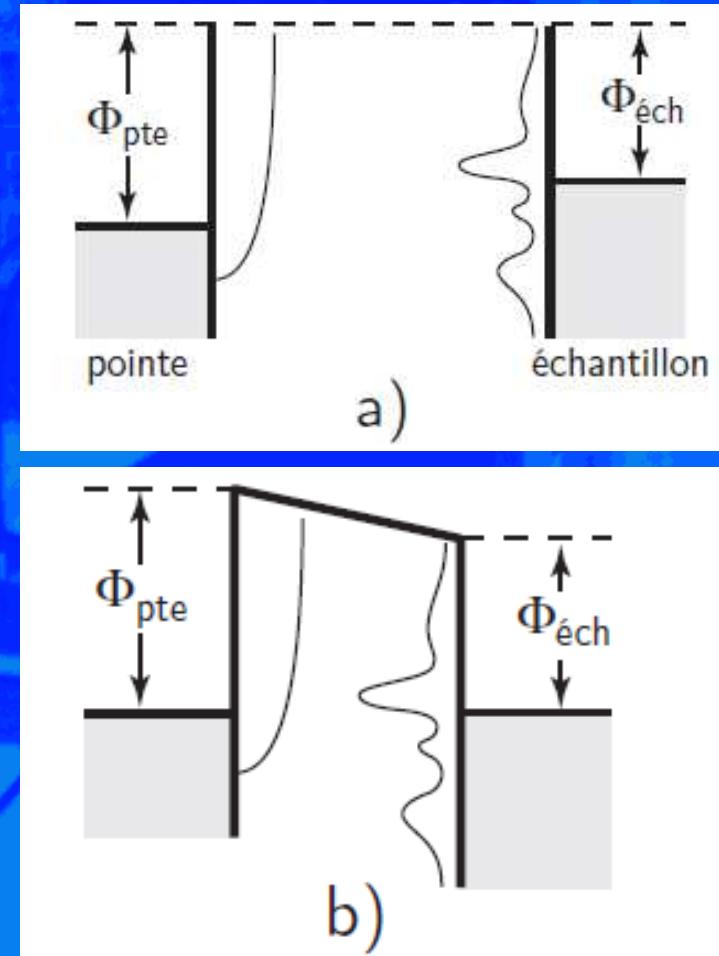
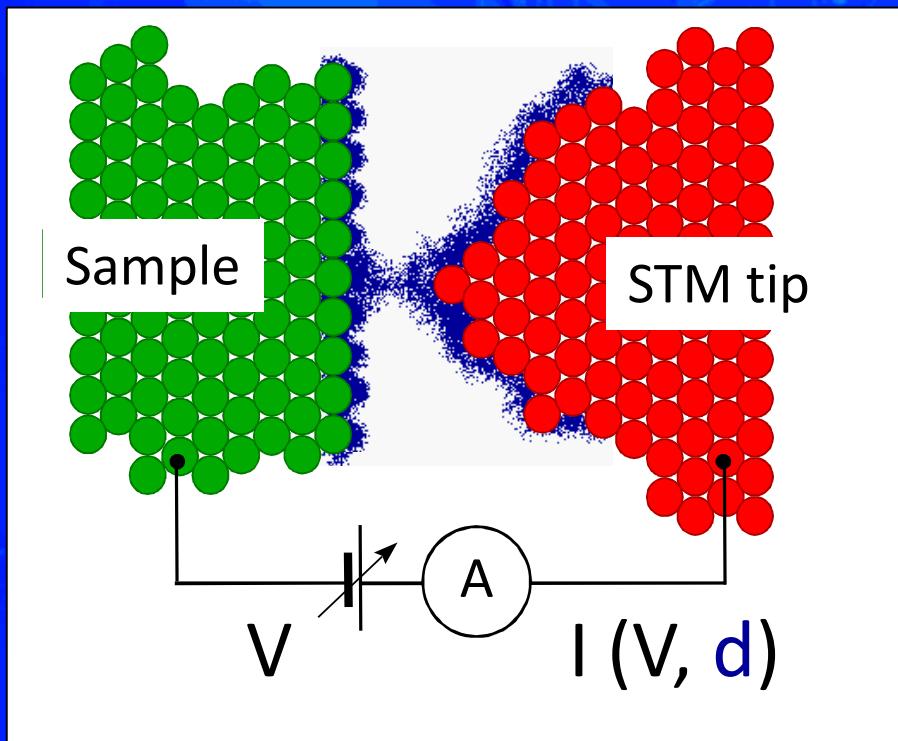
Ecole MICO 2010

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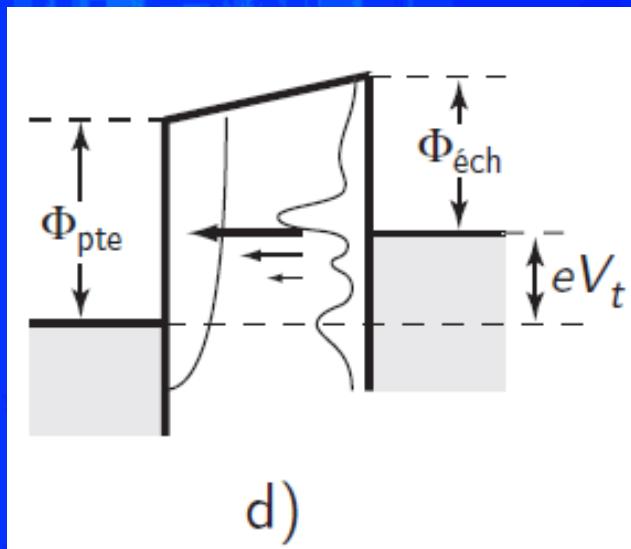
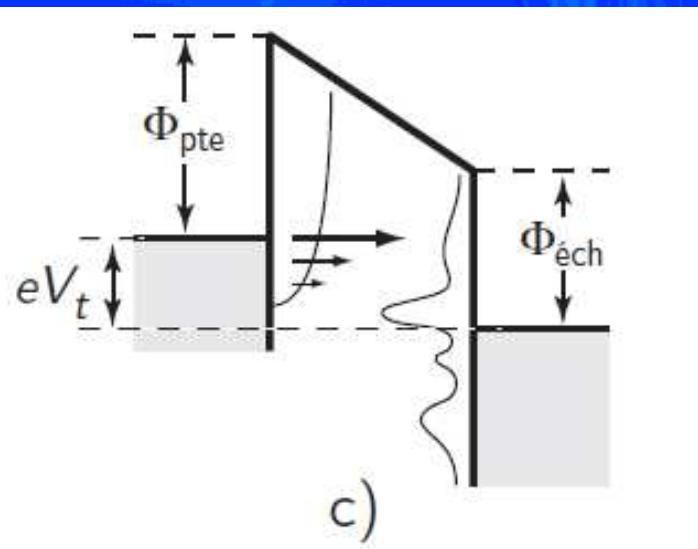
1. Physical background
2. How to make it, how it looks like
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STM/STS: Local Tunneling Junction



N.B. STM junction =
Sample + Tip + Cable + Battery (+ Ampermeter)

Tunneling Junction at T > 0K



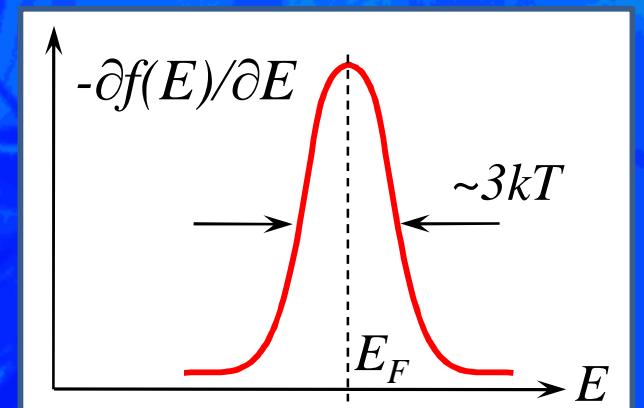
$$I = \frac{2\pi e}{\hbar} \int_{-\infty}^{\infty} TN_s(E - eV)N_p(E)[f(E - eV) - f(E)]dE$$

If $N_p(E), T(E) = \text{const}$

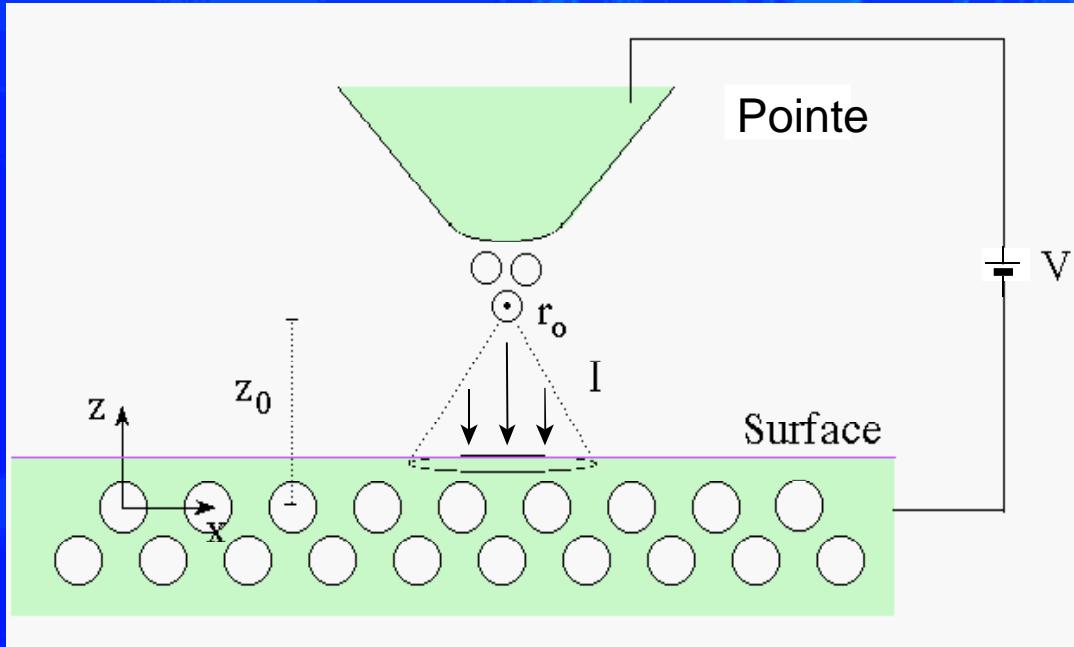
$$\frac{dI}{dV}(V) = -\frac{2\pi e^2 T}{\hbar} N_p \int_{-\infty}^{\infty} N_s(E + eV) \frac{\partial f}{\partial E}(E)dE$$

At zero Temperature $T = 0$:

$$\frac{dI}{dV}(V) = \frac{2\pi e^2 T}{\hbar} N_p N_s(eV) !$$



Local Tunneling Junction at T > 0K



- J. Tersoff et D. R. Hamann, Phys. Rev. Lett. 50, 1998 (1983).
A. Selloni, et al. Phys. Rev. B 31, 2602 (1985).
N. Lang, Phys. Rev. B 34, 5947 (1986).

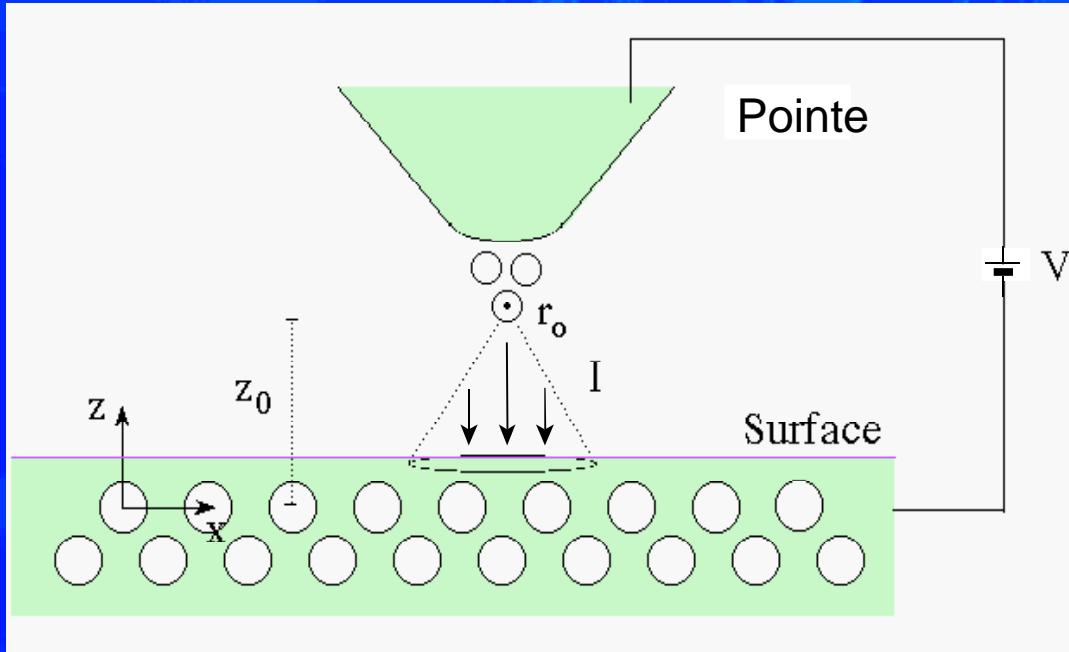
$$I(V, T, x, y, s) \propto \int_{-\infty}^{\infty} dE \rho_{\text{éch}}(E, x, y, s) \rho_{\text{pte}}(E - eV) \times T(E, V, s) [f(E - eV, T) - f(E, T)]$$

$$\rho_{\text{éch}}(E, x, y, s) = \sum_k |\psi_k(\mathbf{r})|^2 \delta(E - E_k)$$

LDOS
Local Density of States

$$T(E, V, s) = \exp \left(-2s \sqrt{\frac{m_e}{\hbar^2}} \sqrt{\Phi_{\text{éch}} + \Phi_{\text{pte}} - 2E + eV} \right)$$

Local Tunneling Junction at T > 0K



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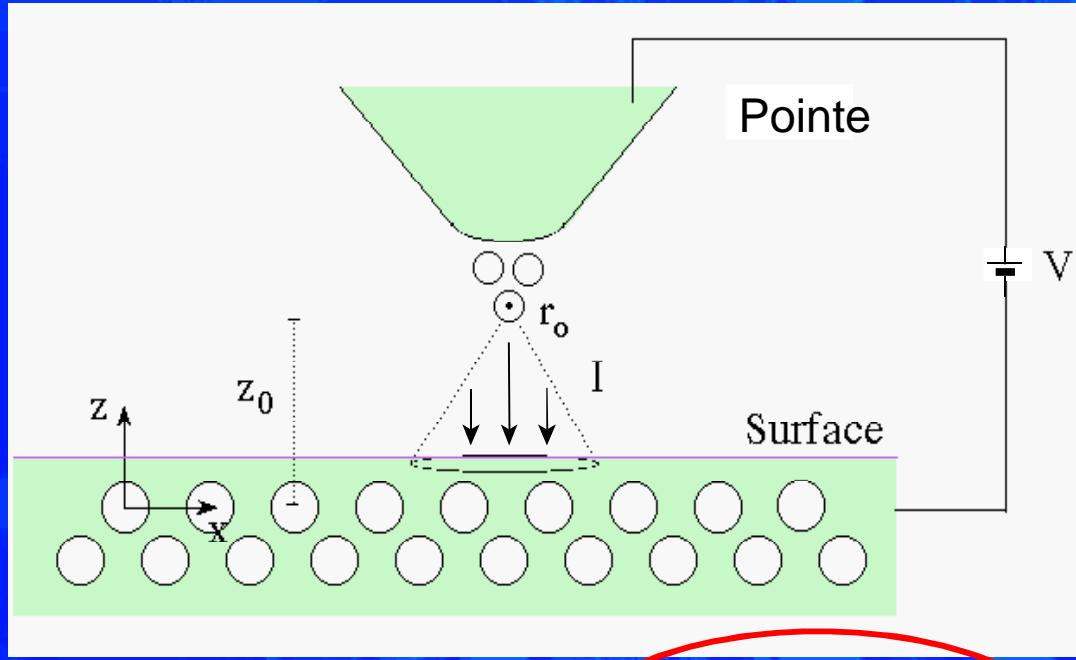
$$I(V, T, x, y, s) \propto e^{-2s\sqrt{\frac{2m_e\Phi}{\hbar^2}}} \times \int_{-\infty}^{\infty} dE \rho_{\text{éch}}(E, x, y) \rho_{\text{pte}}(E - eV) g(E, V, T)$$

where $g(E, V, T) = f(E - eV, T) - f(E, T)$

If $T = 0$: $\frac{dI}{dV}(V) = -\frac{2\pi e^2 T}{\hbar} N_p N_s(eV)$

$\rho(\mathbf{r}, E) = \sum_k |\psi_k(\mathbf{r})|^2 \delta(E - E_k)$

Local Tunneling Junction at T > 0K



J. Tersoff et D. R. Hamann, Phys. Rev. Lett. 50, 1998 (1983).
 A. Selloni, et al., Phys. Rev. B 31, 2602 (1985)
 N. I. Smith, et al., Phys. Rev. B 34, 5947 (1986).

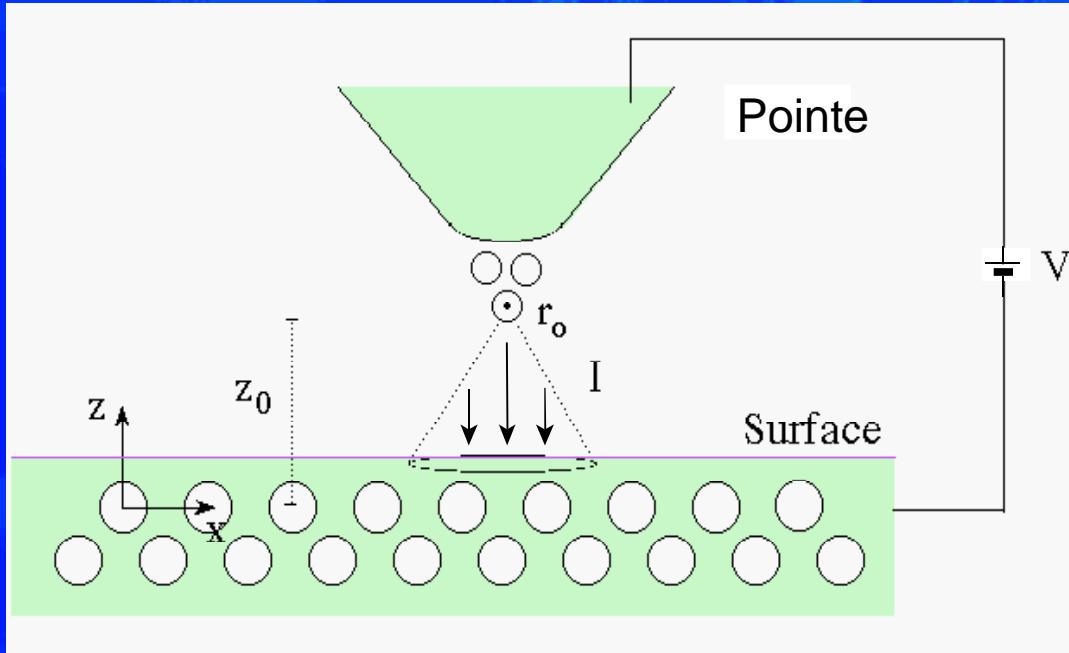
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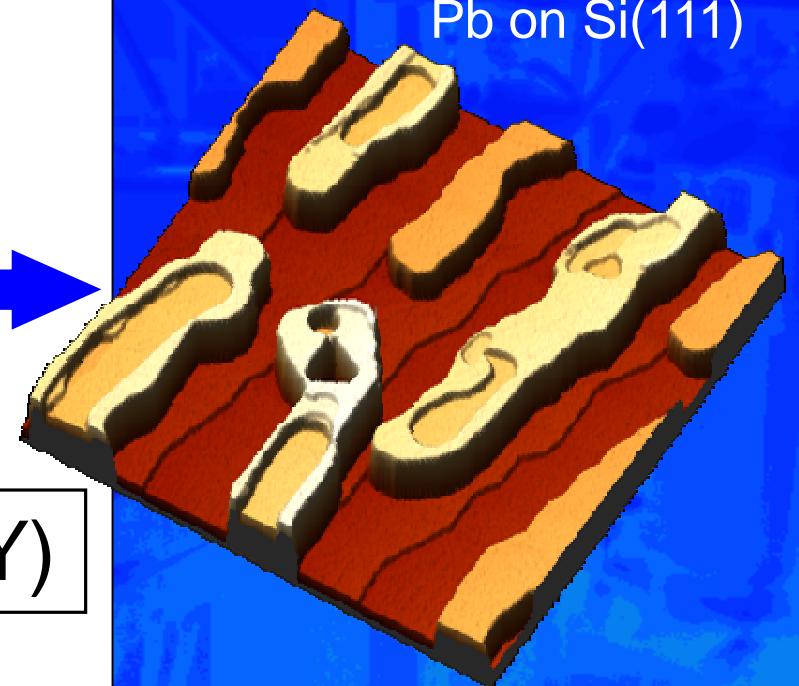
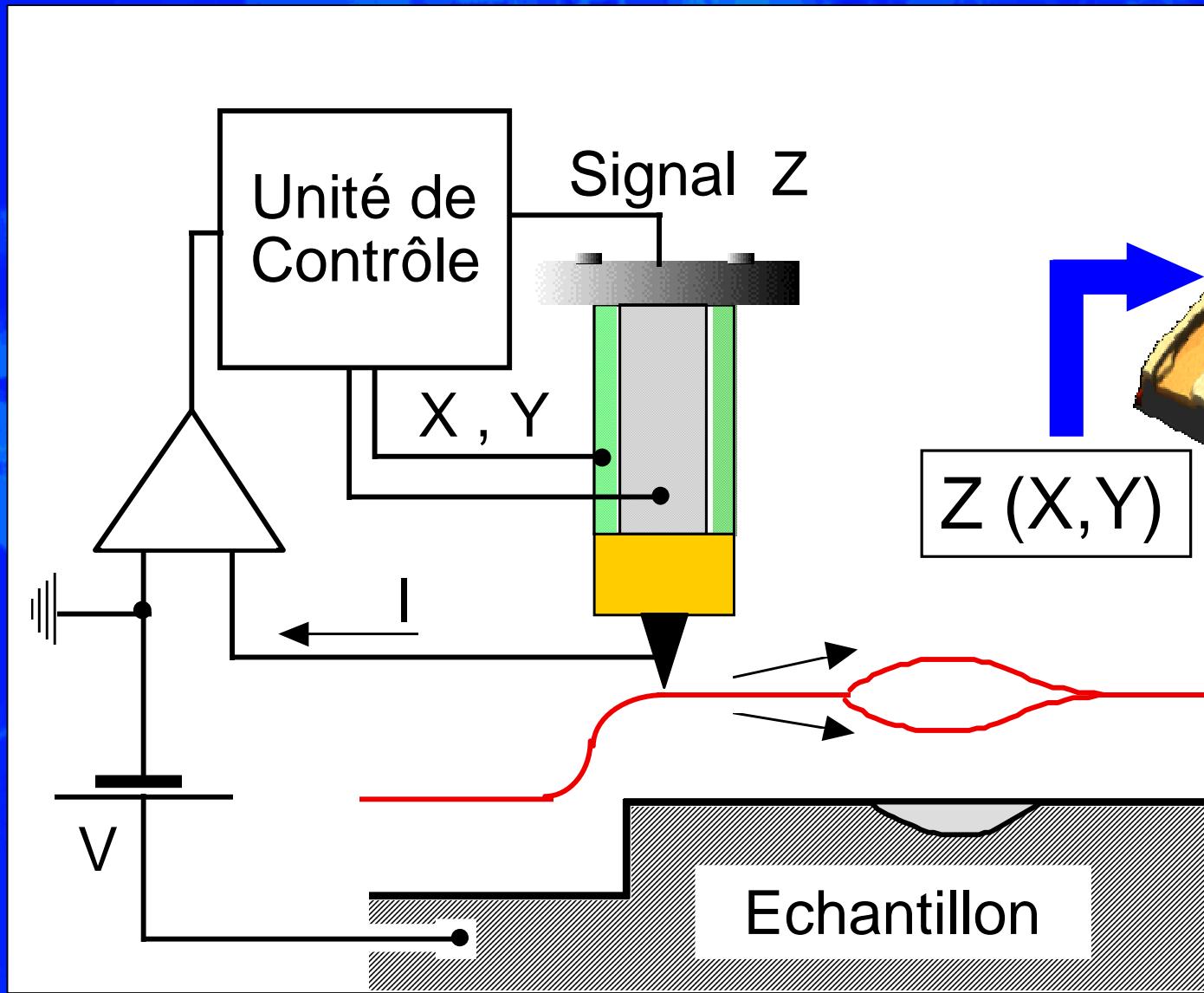
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where $g(E, V, T) = f(E - eV, T) - f(E, T)$

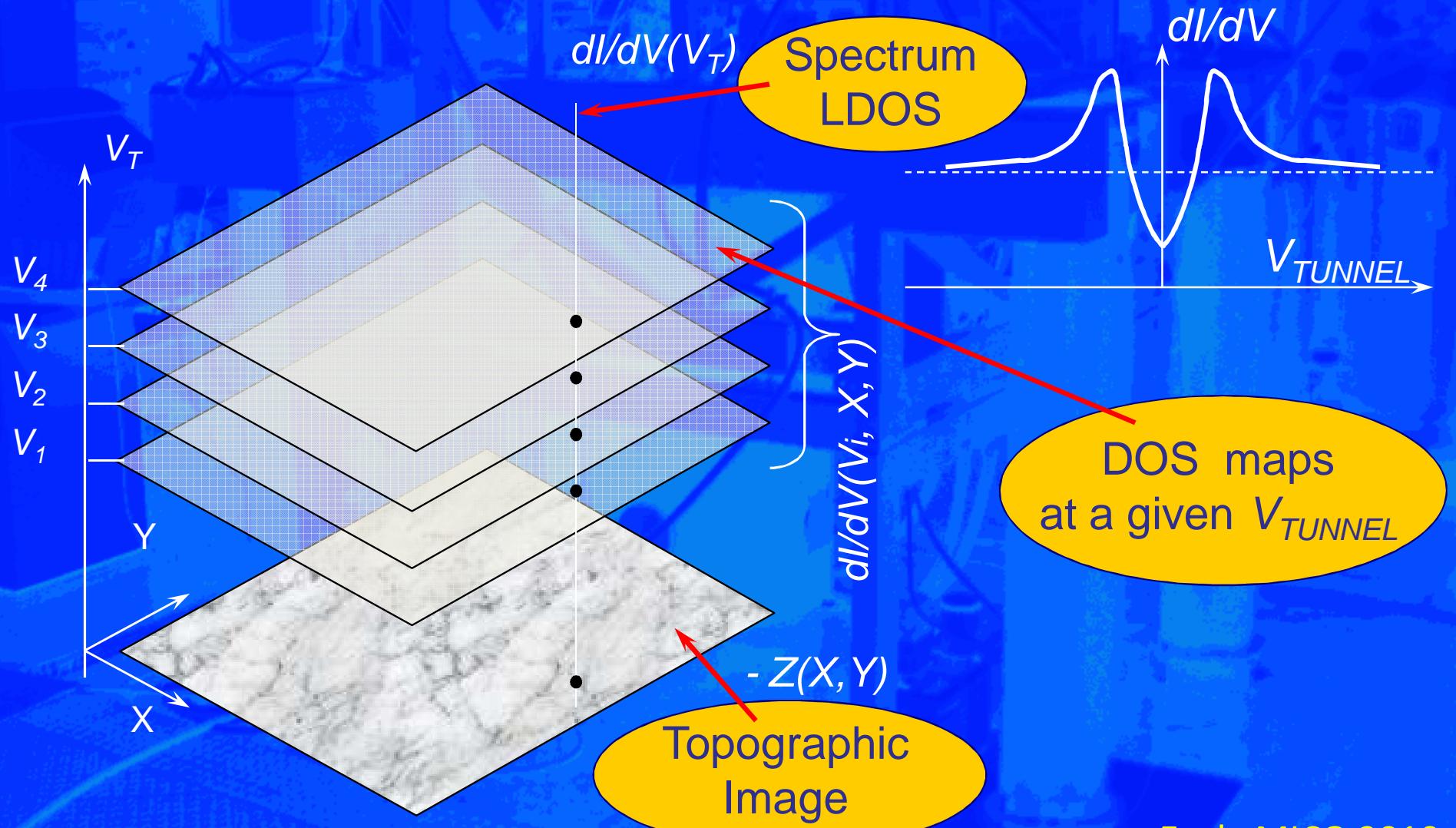
If $T = 0$: $\frac{dI}{dV}(V) = -\frac{2\pi e^2 T}{\hbar} N_P N_S(eV)$

$$\rho(\mathbf{r}, E) = \sum_k |\psi_k(\mathbf{r})|^2 \delta(E - E_k)$$

STM: Working principle



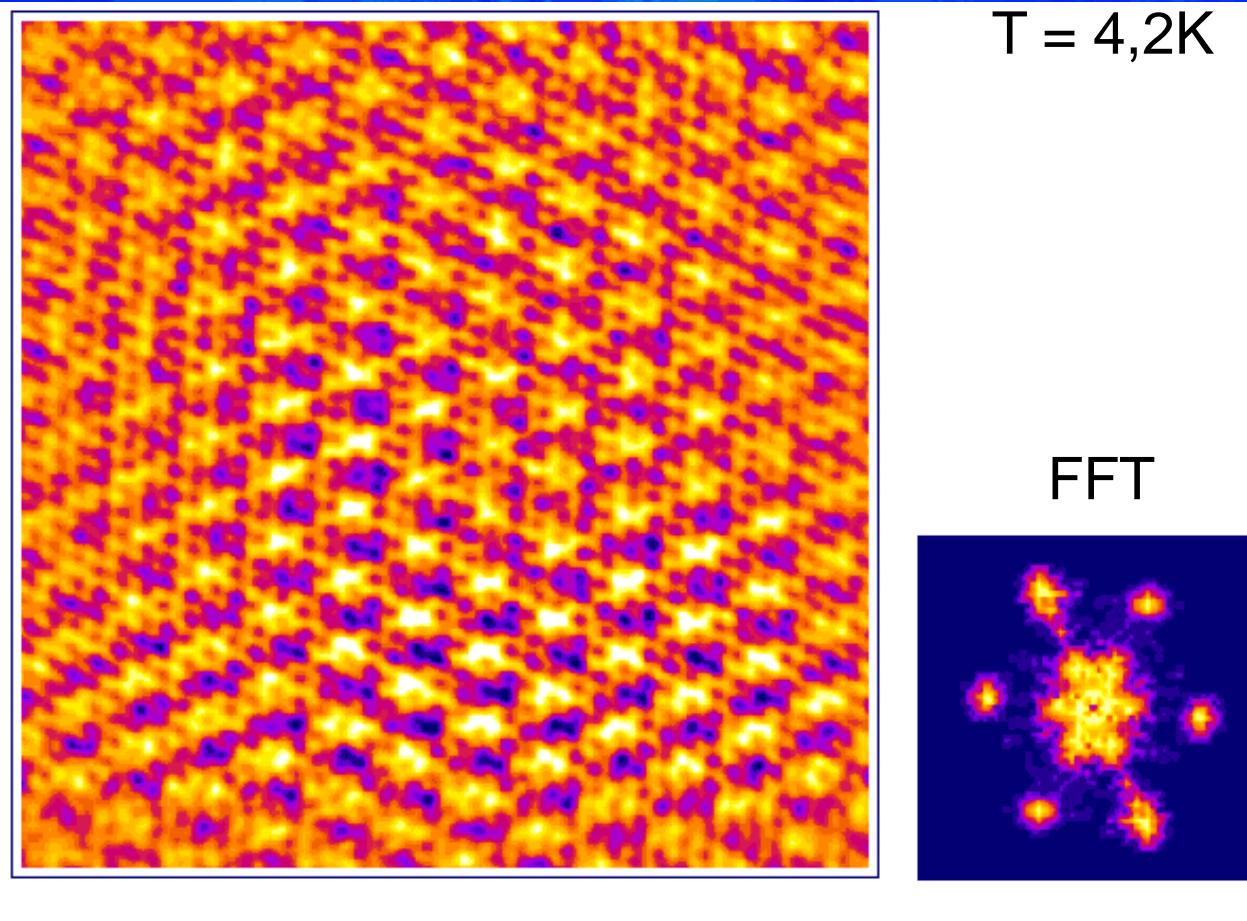
STM/STS : Scanning Tunneling Spectroscopy



STS: Charge Density Waves in NbSe₂

T_ODC = 33K

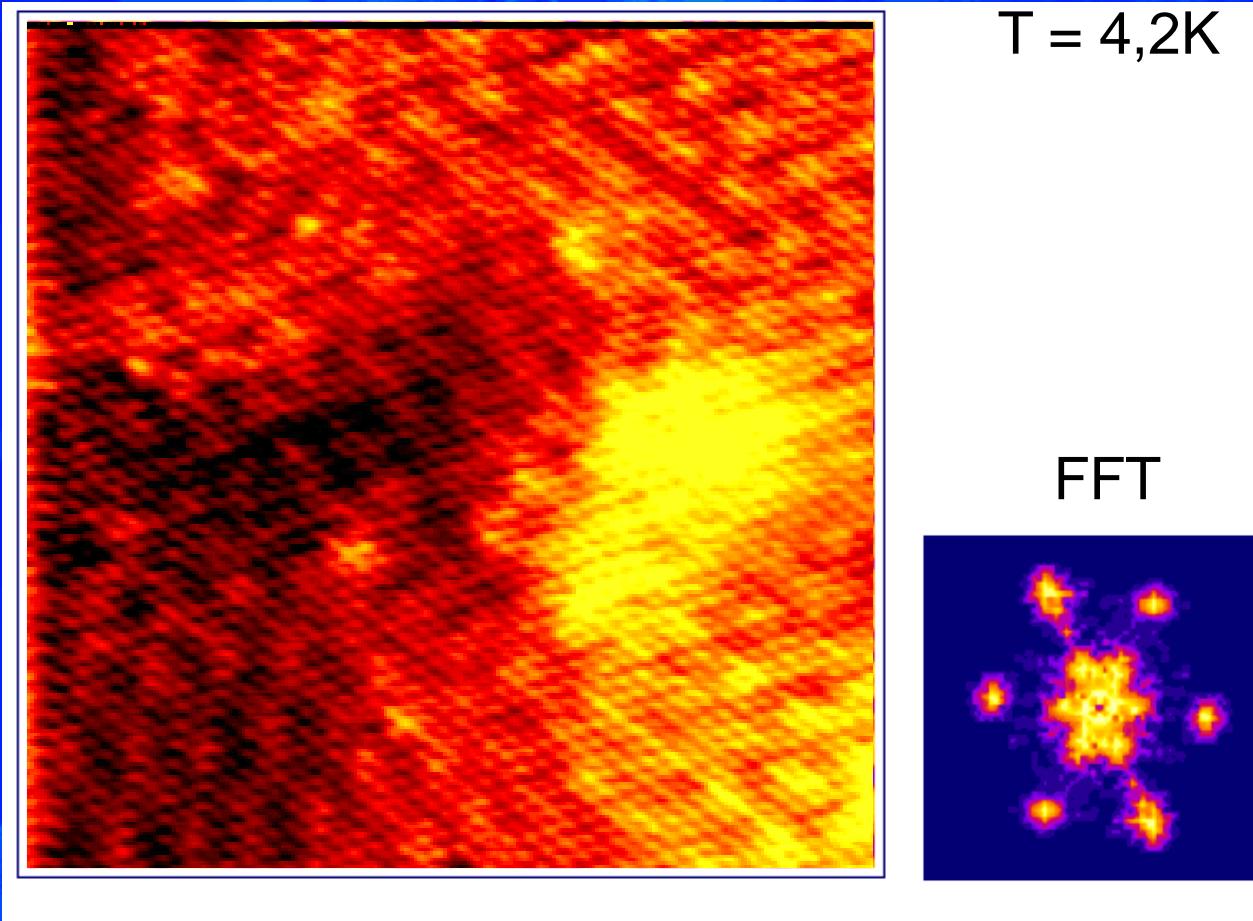
Topographic STM Image 13nm x 13 nm



STS: Charge Density Waves in NbSe₂

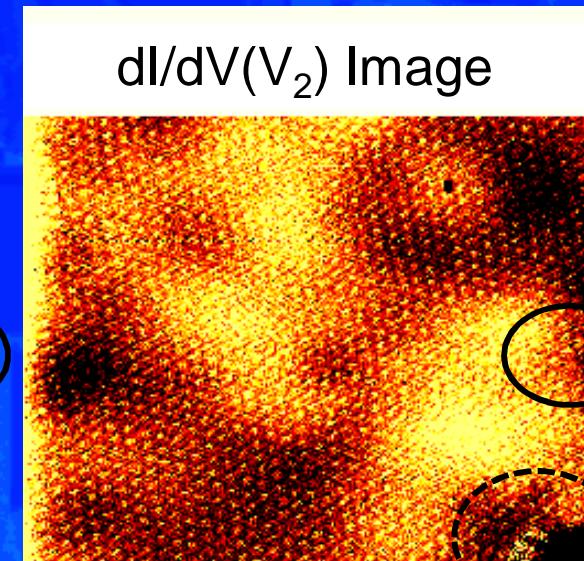
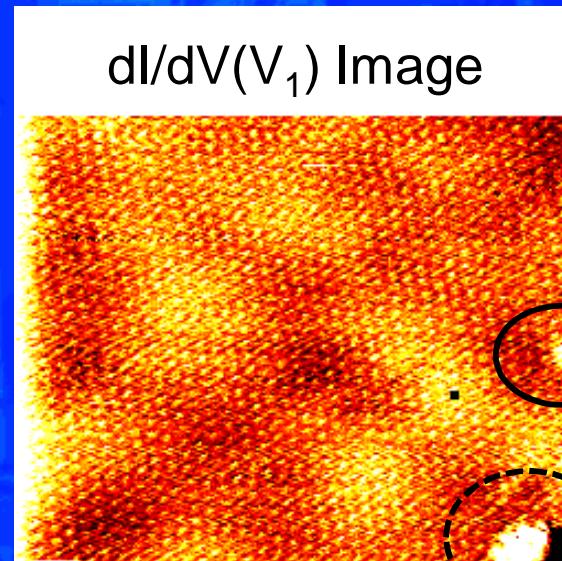
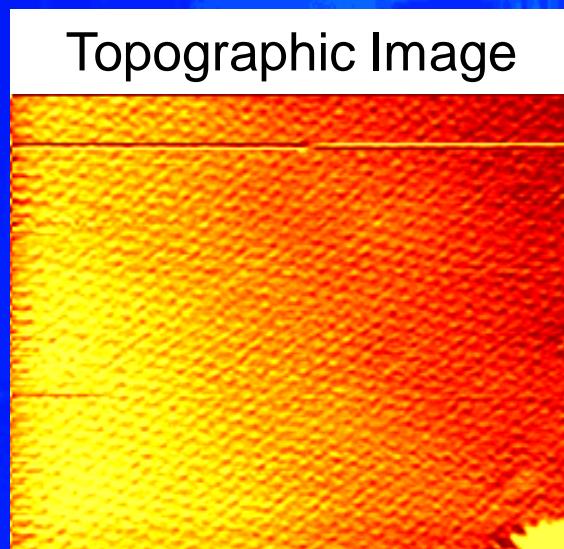
T_{ODC} = 33K

Topographic STM Image 18nm x 18 nm



STS: Charge Density Waves in NbSe₂

STM/STS



T. Cren, D. Roditchev, W. Sacks (1995-2000)

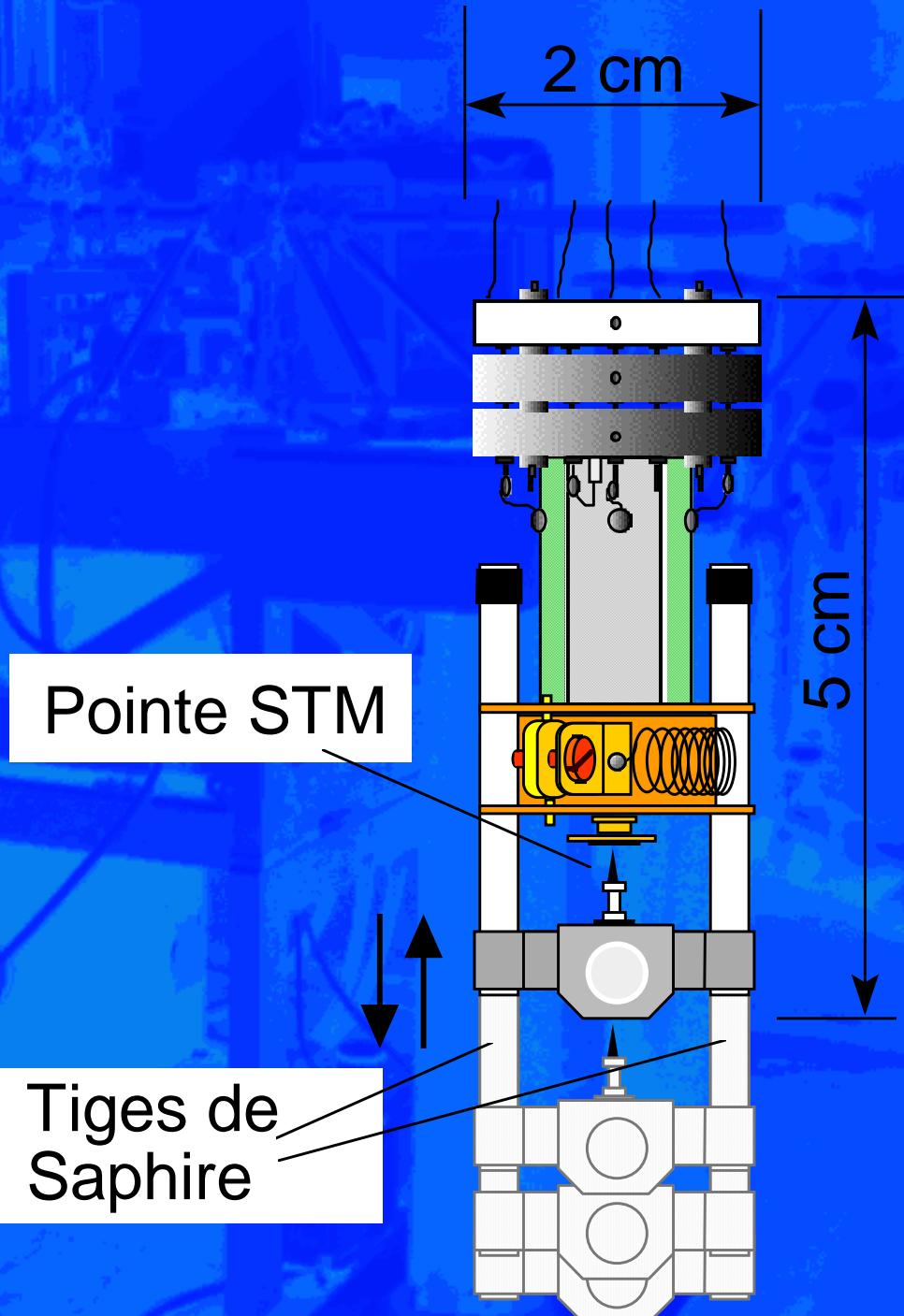
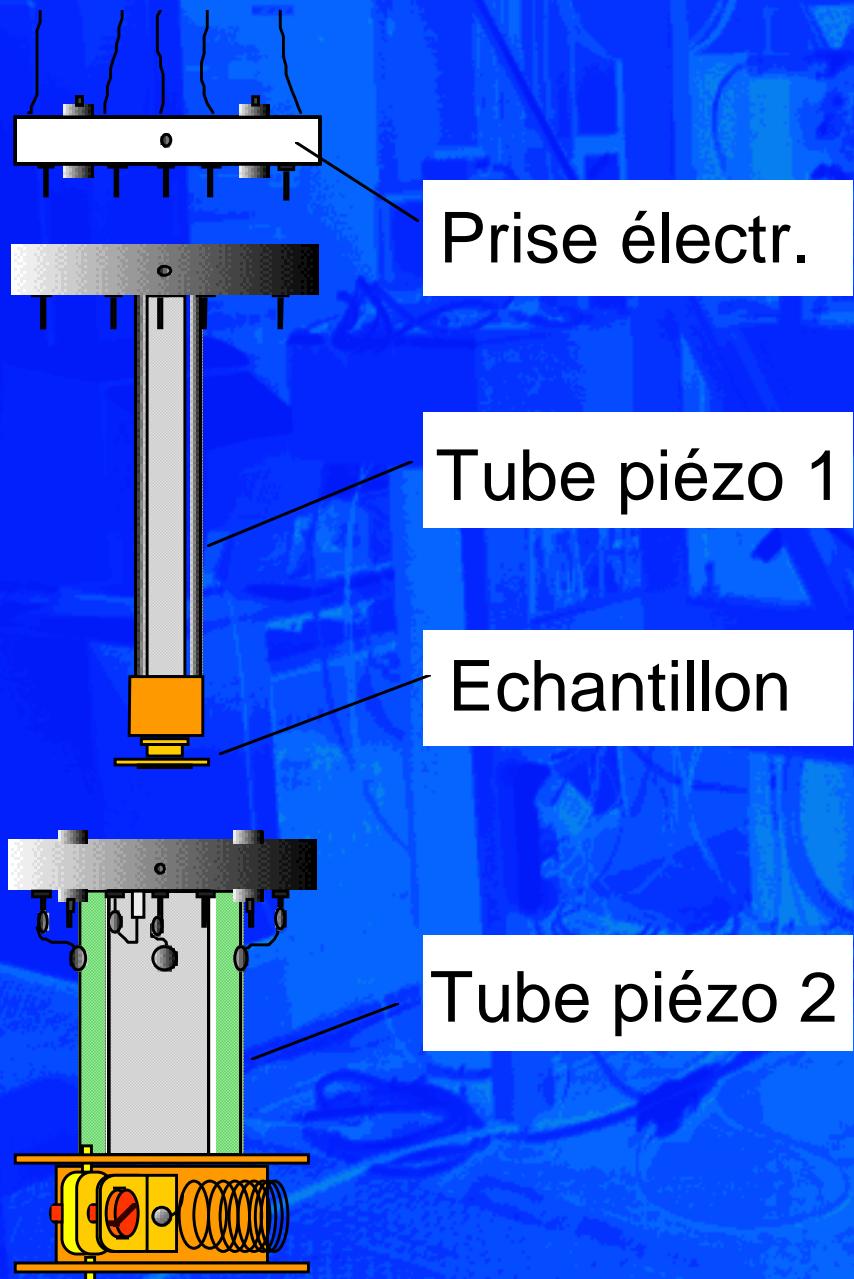
N.B. Charge Density Waves are strongly perturbed by local disorder (the defects lying under surface are detectable !)

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Microscopes à effet tunnel



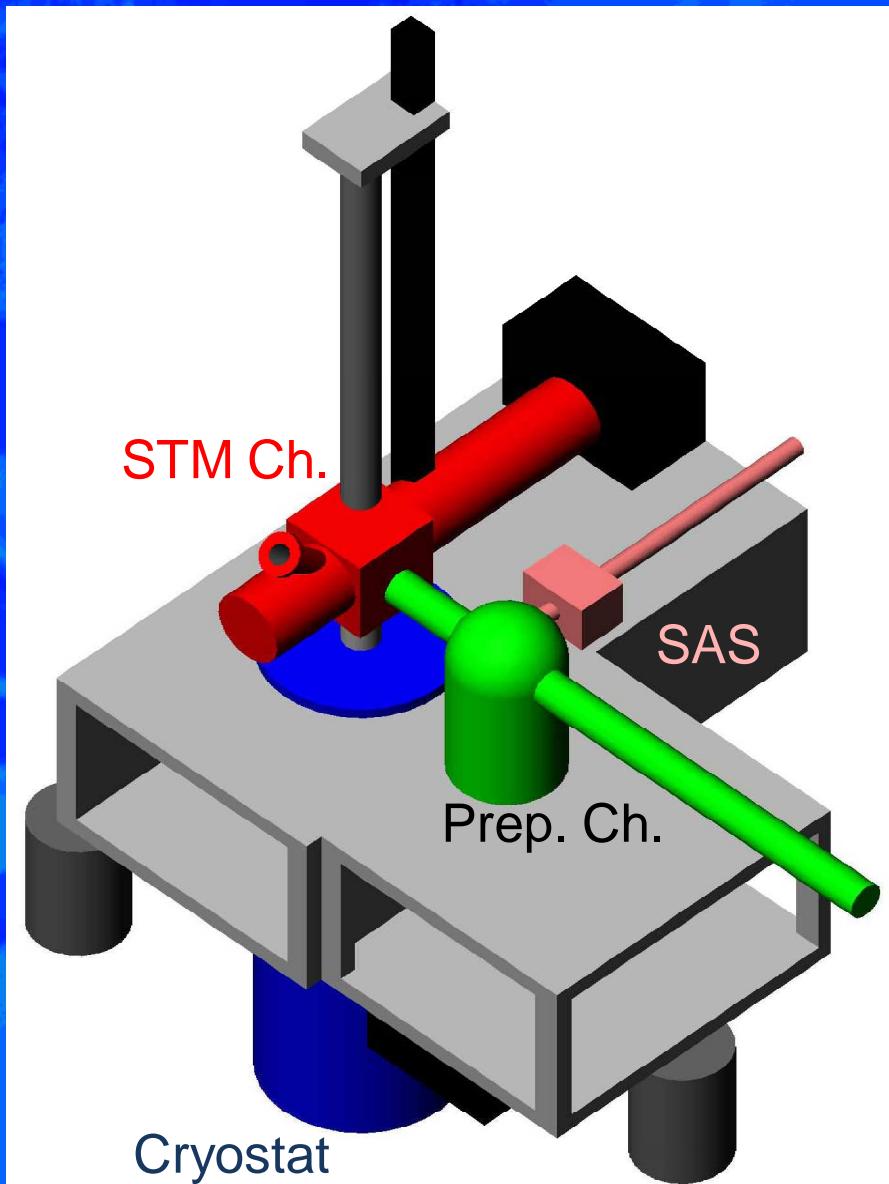
Microscope tunnel fonctionnant à l'air

« Easy-Scan »

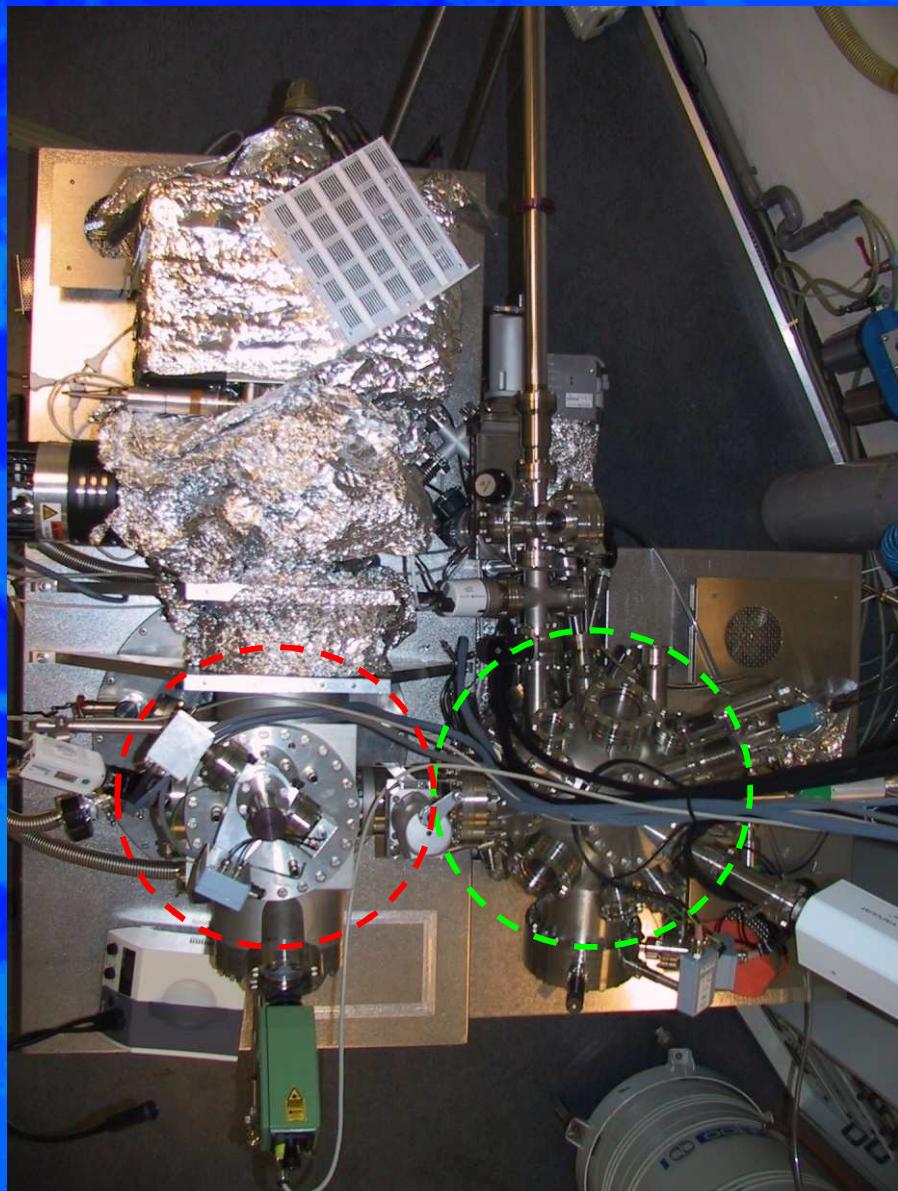
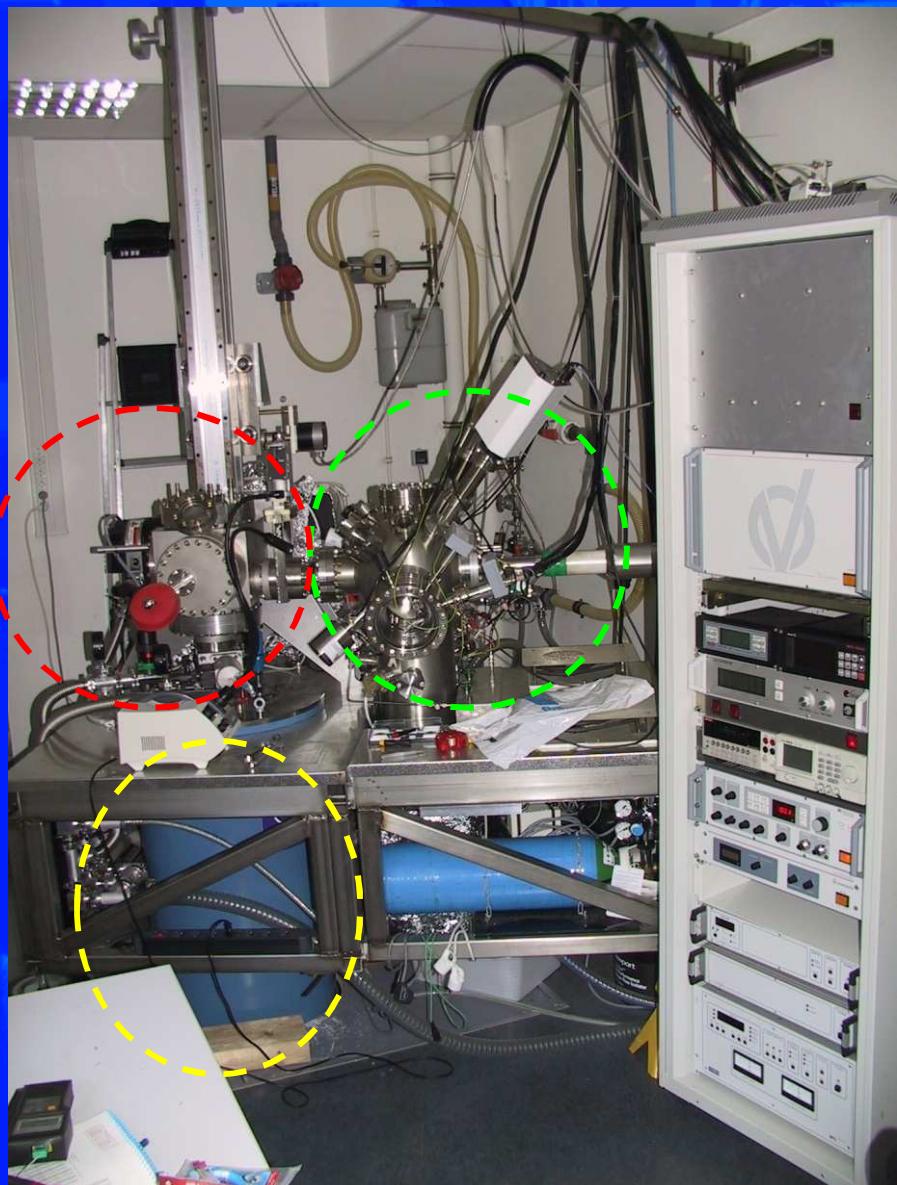


commercialisé par la société Schaefer Techniques, prix <10.000 E

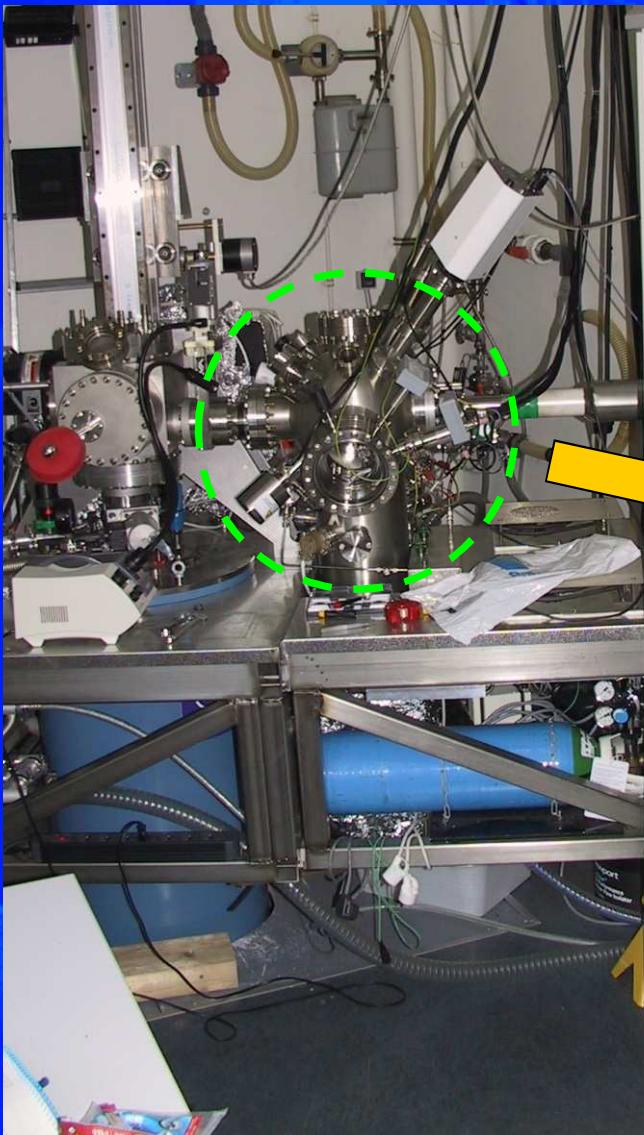
Scanning Tunneling Microscopy and Spectroscopy at INSP in Paris



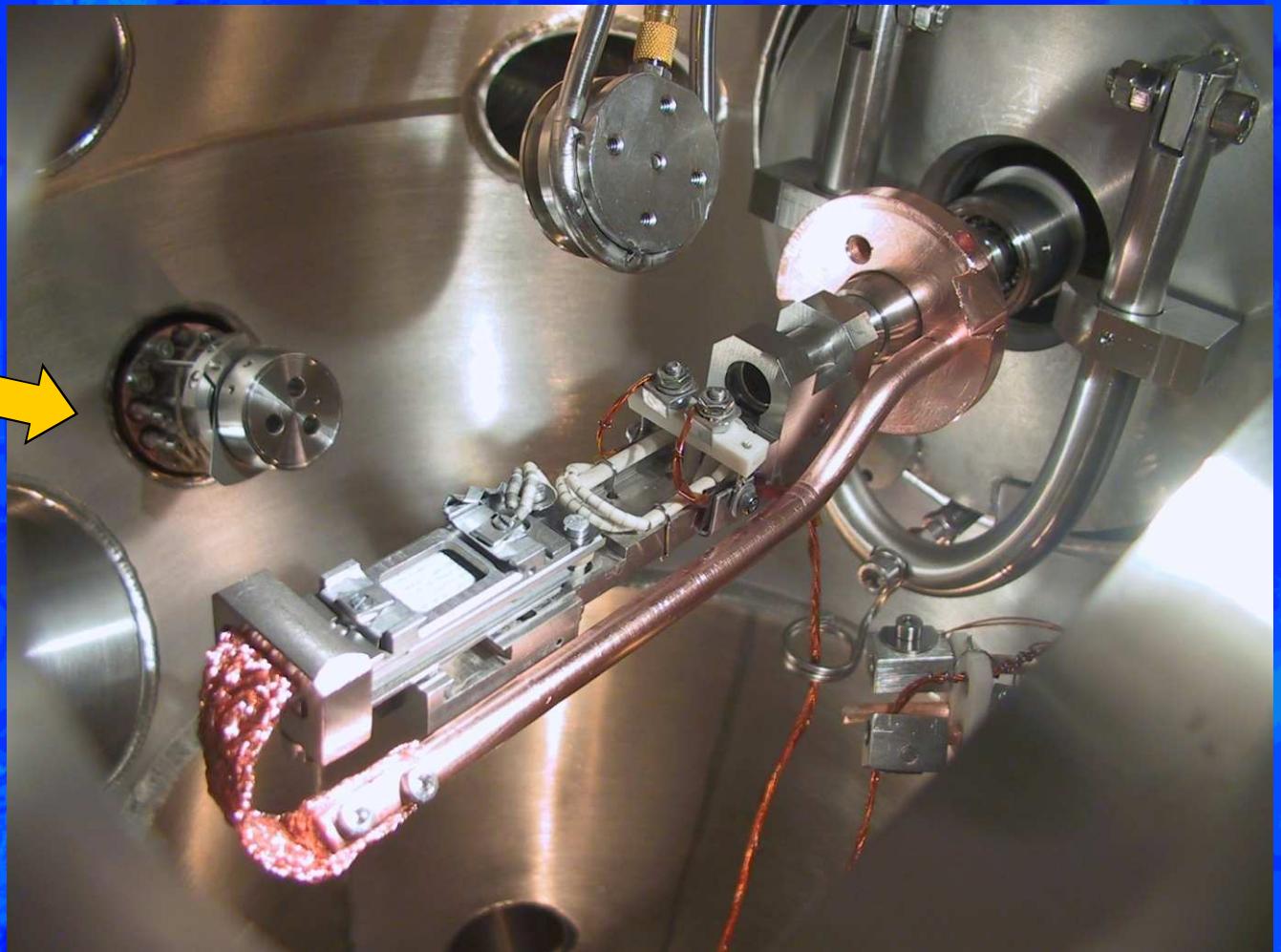
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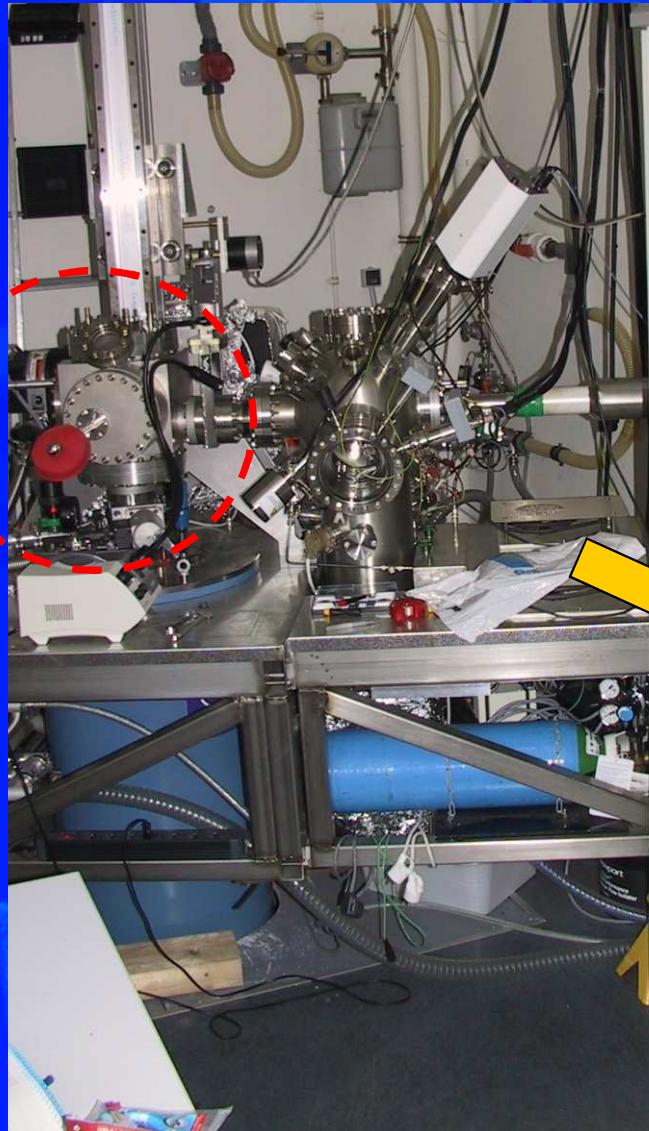
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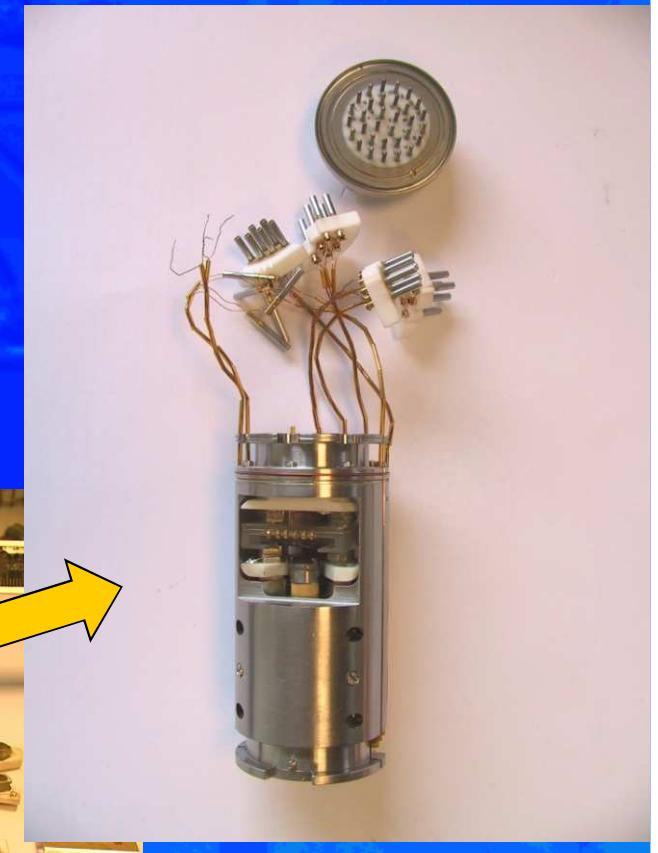
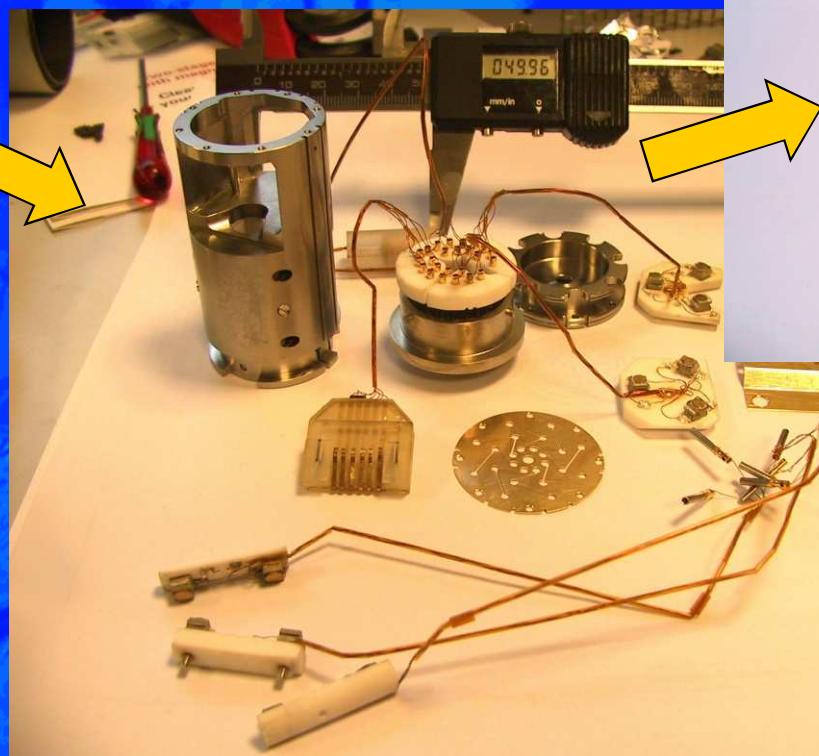
Electron Beam Deposition under UHV



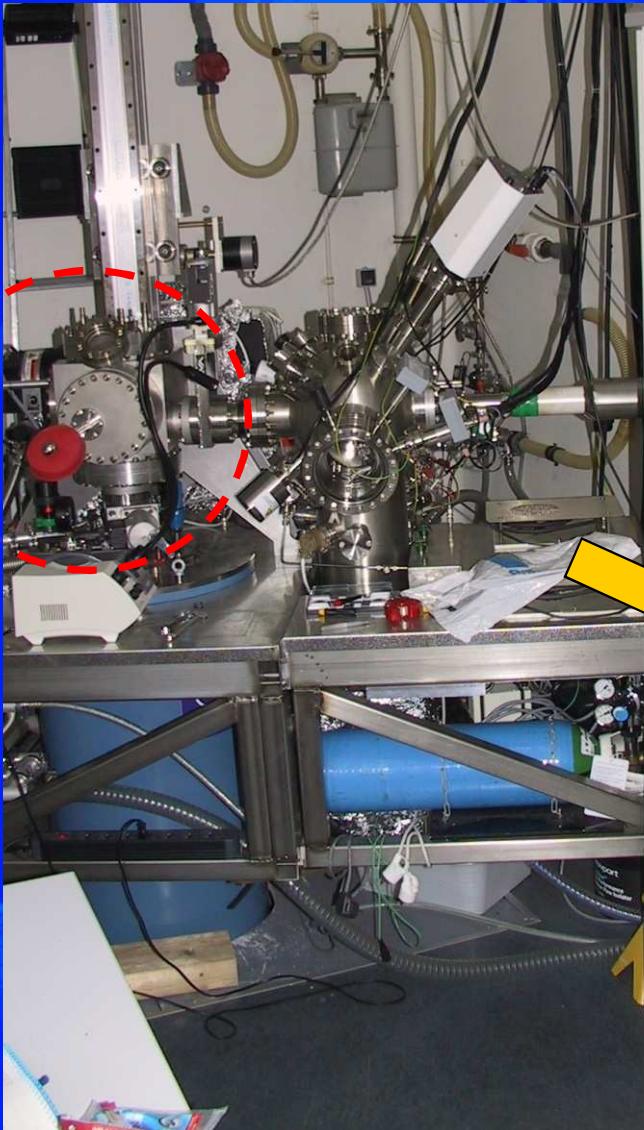
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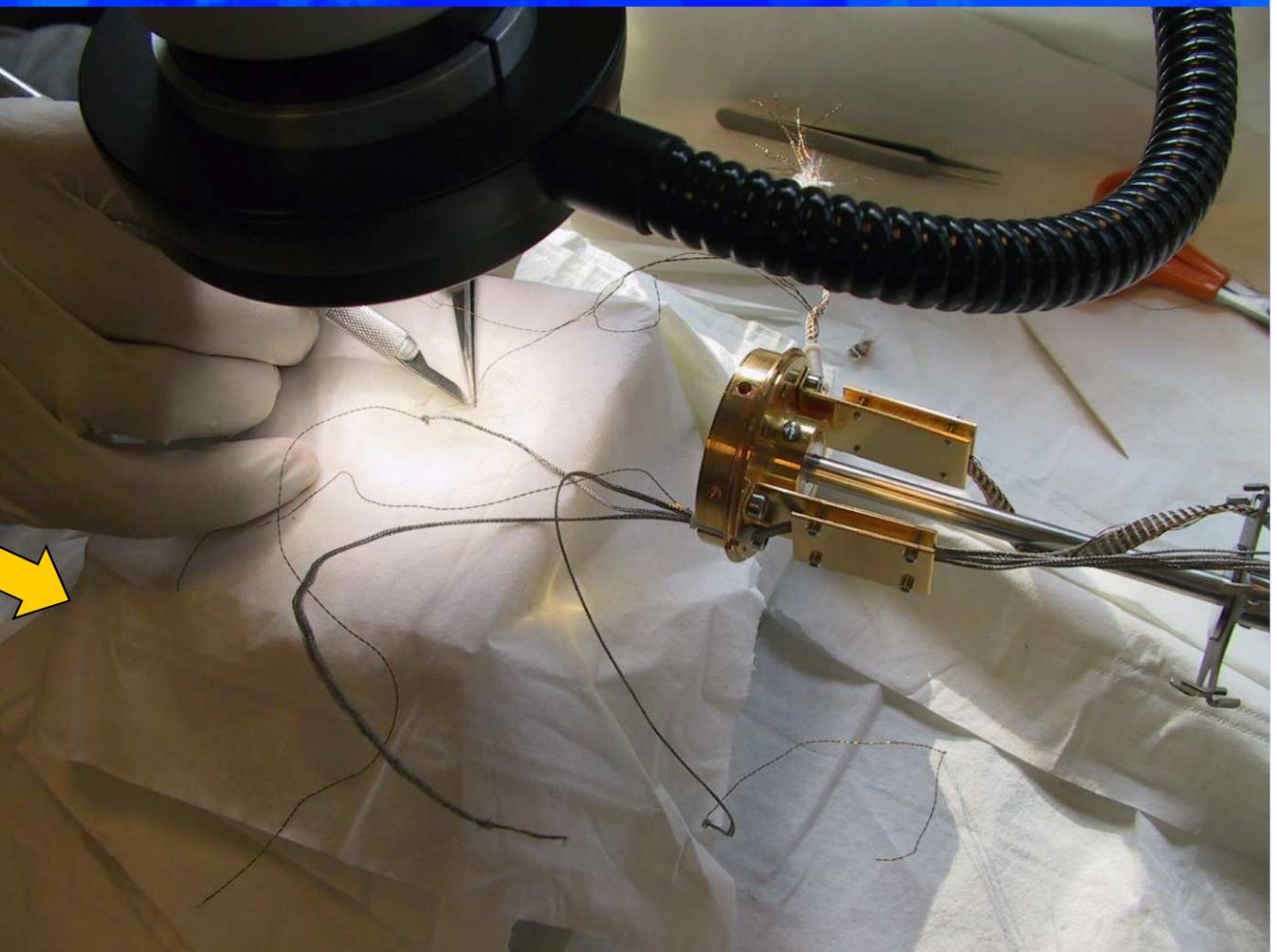
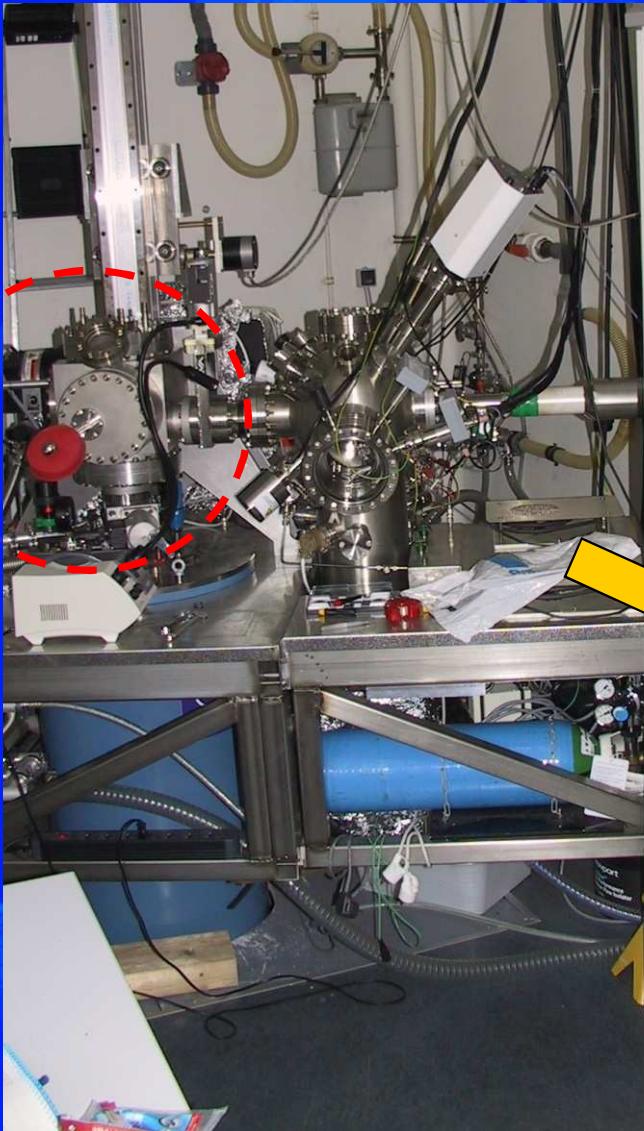
A new STM Head



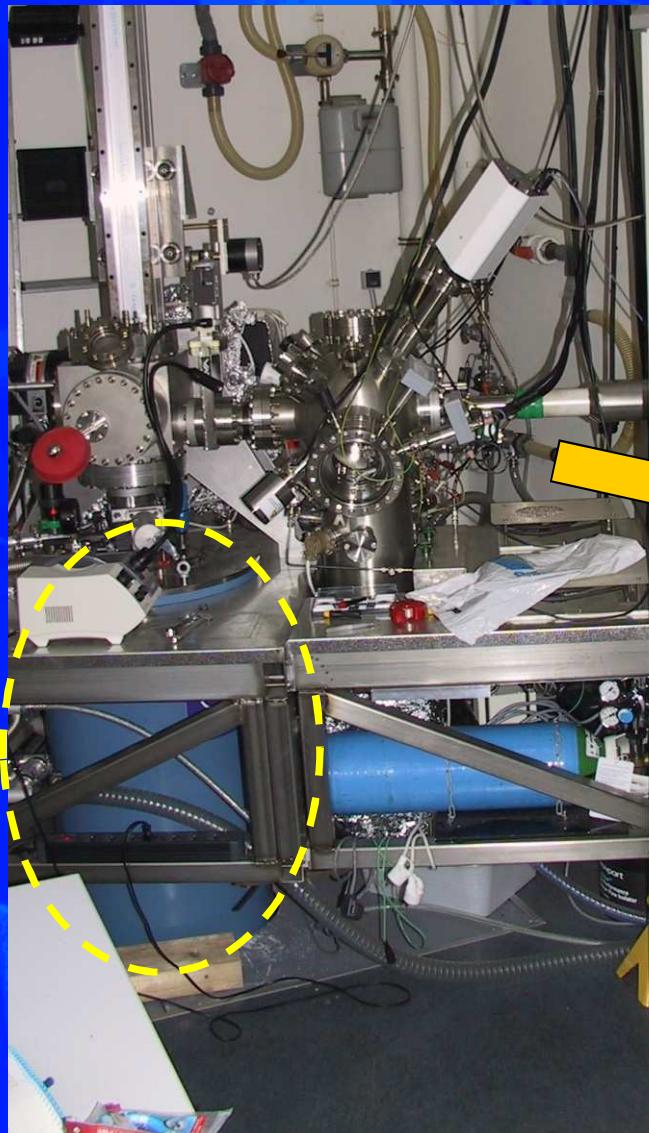
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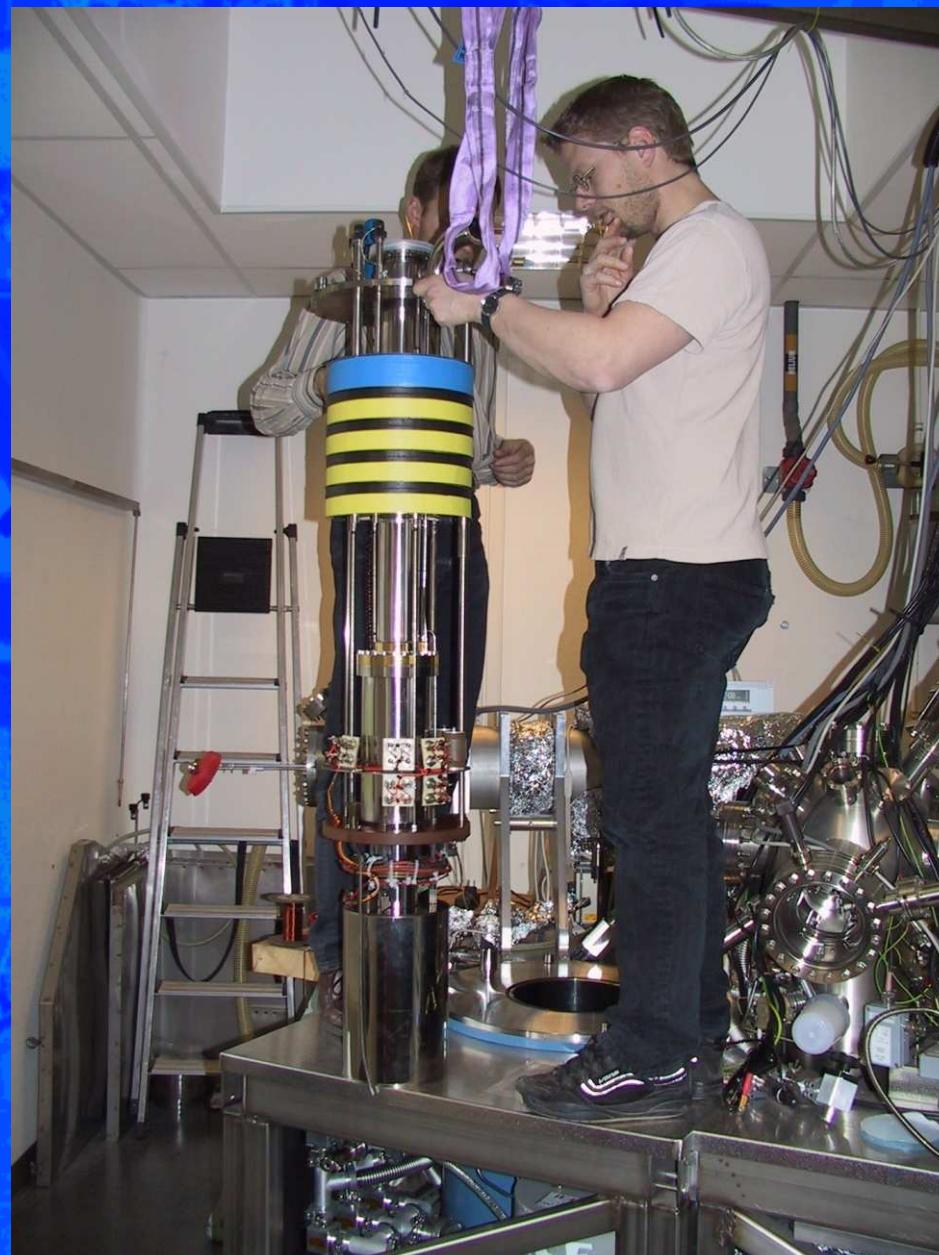
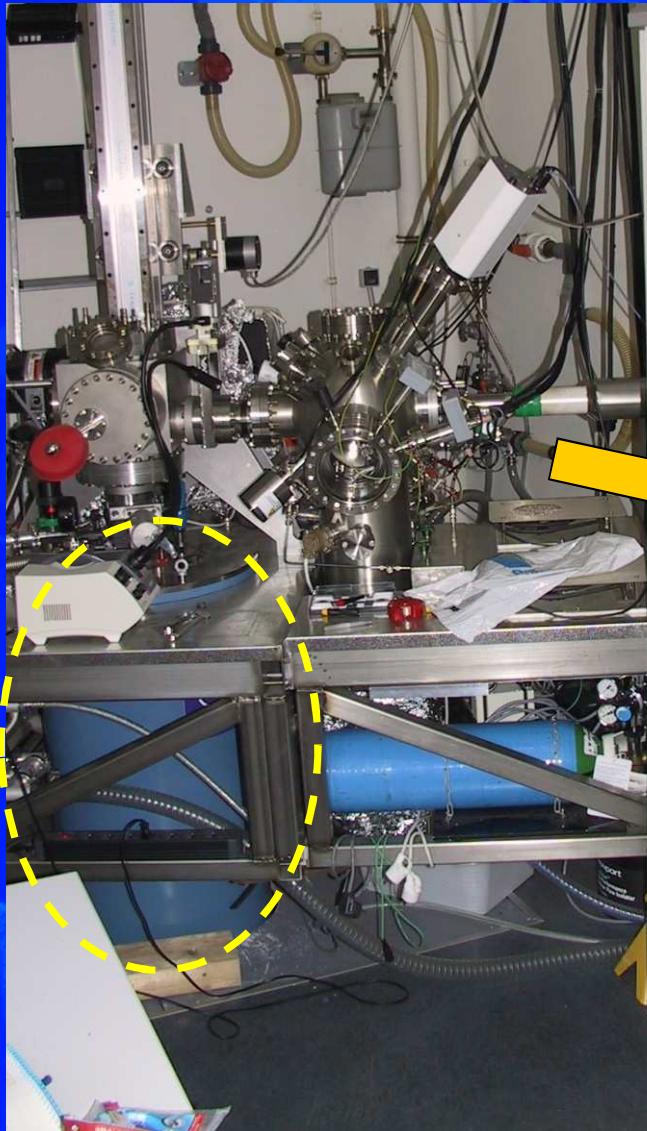


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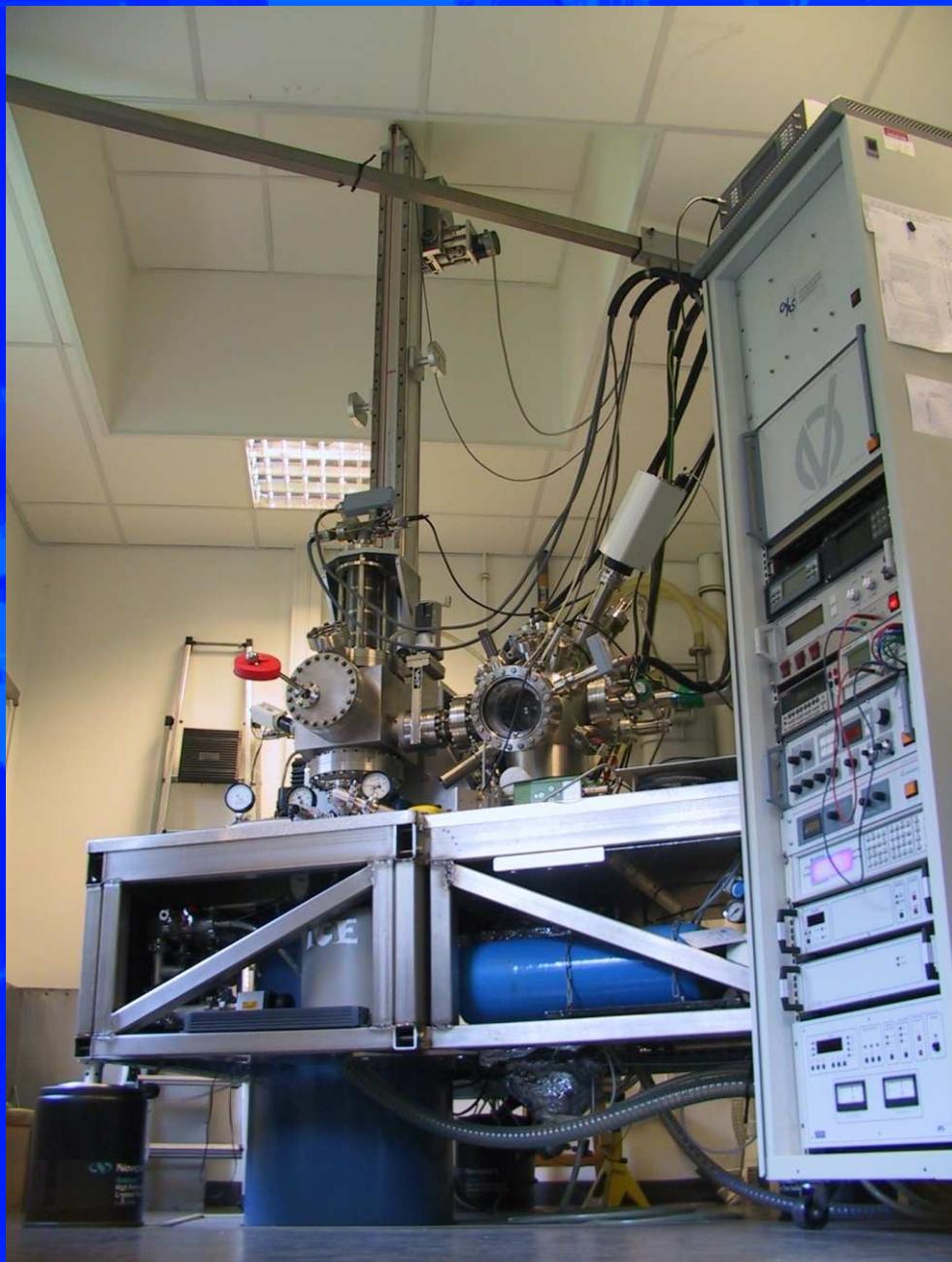


Ultra-Low Temperatures 0.3K
Magnetic Field up to 10Tesla

Scanning Tunneling Microscopy and Spectroscopy at INSP in Paris



Scanning Tunneling Microscopy and Spectroscopy at INSP in Paris



Ultra-Low Temperatures 0.3K
Magnetic Field up to 10Tesla



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Superconductivity: Ginzburg-Landau Approach

Superconducting phase is described by macroscopic wave function:

$$\Psi = |\Psi| \exp(i\varphi)$$

$$n_s = |\Psi(\vec{r})|^2$$

Two equations:

$$\frac{1}{2m^*}(-i\hbar\vec{\nabla} - 2e\vec{A})^2\Psi + \beta|\Psi|^2\Psi = -\alpha\Psi \quad (1)$$

$$\vec{j} = \frac{2e}{m^*}|\Psi|^2(\hbar\vec{\nabla}\varphi - 2e\vec{A}) = 2e|\Psi|^2\vec{v}_s \quad (2)$$

where $\vec{v}_s = (\hbar\vec{\nabla}\varphi - 2e\vec{A})/m^*$

Boundary condition at the sample edge:

$$(-i\hbar\vec{\nabla} - 2e\vec{A})\Psi\Big|_{\perp} = 0.$$

Superconductivity: Ginzburg-Landau Approach

Integrating the 2nd G-L equation over an area S:

$$\oint m^* \vec{v}_s \cdot d\vec{s} = \hbar \oint \vec{\nabla} \varphi \cdot d\vec{s} - 2e \oint \vec{A} \cdot d\vec{s}.$$

where $\oint \vec{A} \cdot d\vec{s} = \Phi$, Φ being the magnetic flux crossing S

Condition on the phase φ (since ψ is a single-valued function):

$$\oint \vec{\nabla} \varphi \cdot d\vec{s} = n2\pi.$$

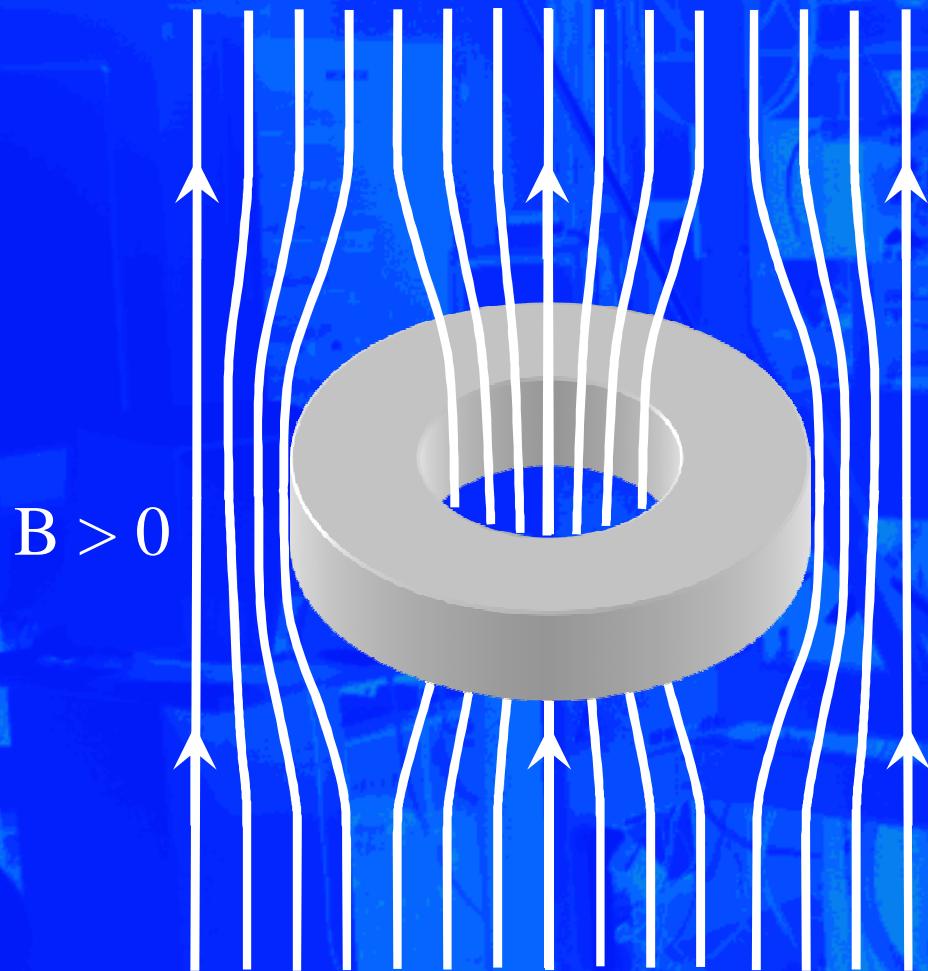
Fluxoid quantification:

$$\Phi' = \Phi + \frac{1}{2e} \oint m^* \vec{v}_s \cdot d\vec{s} = n \frac{h}{2e} = n\Phi_0$$

where Φ_0 is the flux quantum:

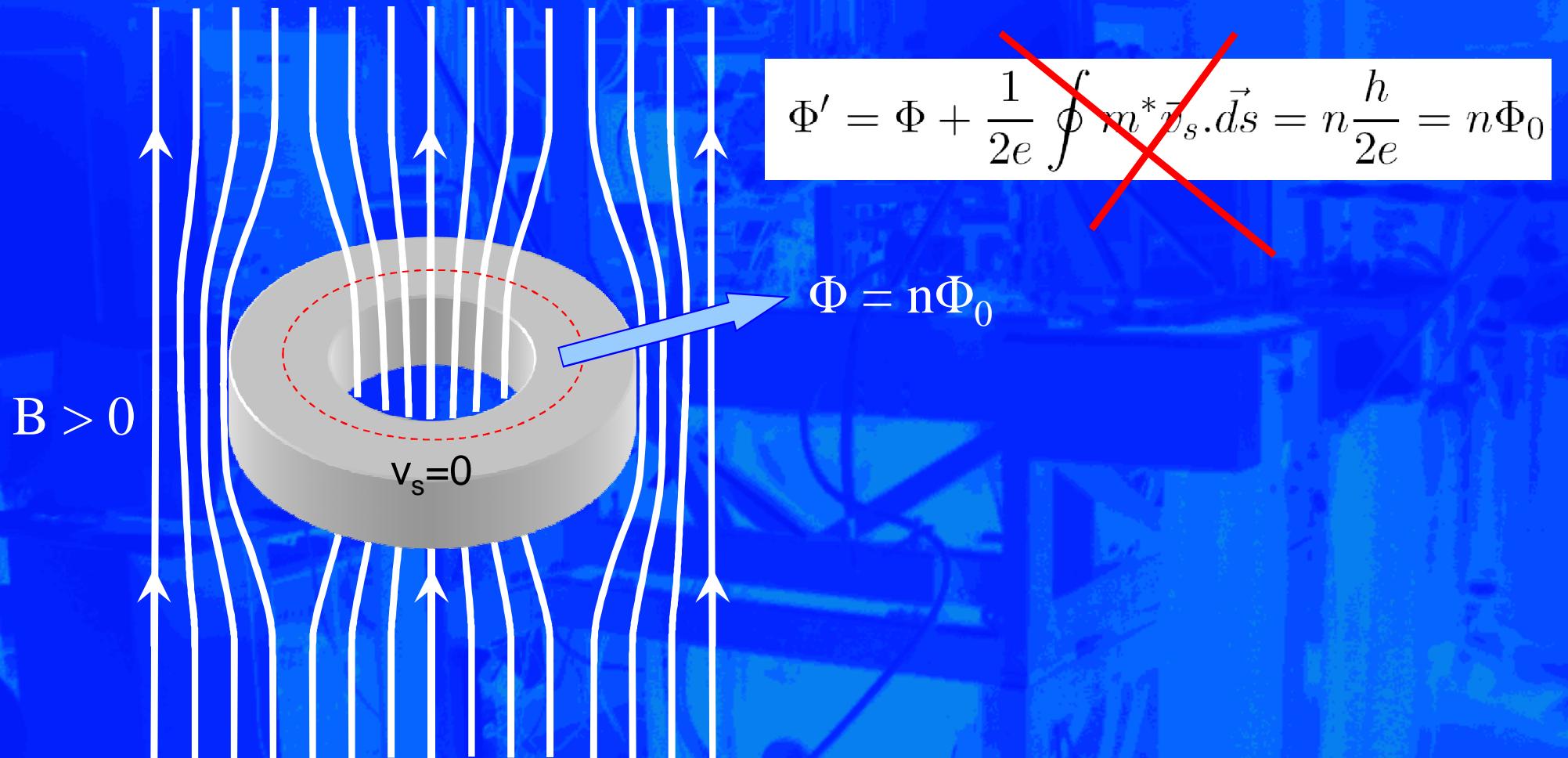
$$\Phi_0 = \frac{h}{2e} = 2.07 \cdot 10^{-15} \text{ Tm}^2$$

Superconductivity: Ginzburg-Landau Approach

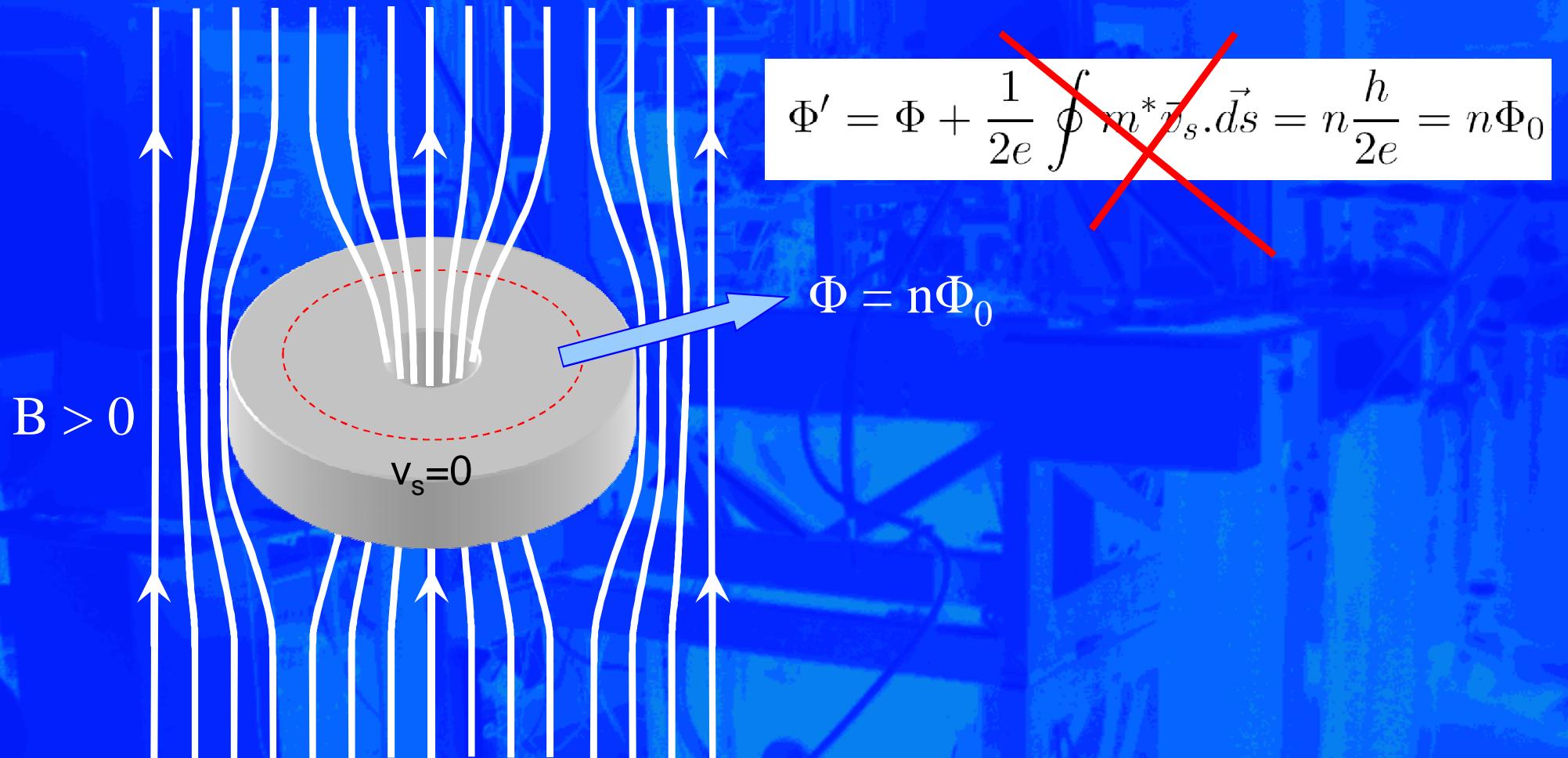


$$\Phi' = \Phi + \frac{1}{2e} \oint m^* \vec{v}_s \cdot d\vec{s} = n \frac{\hbar}{2e} = n\Phi_0$$

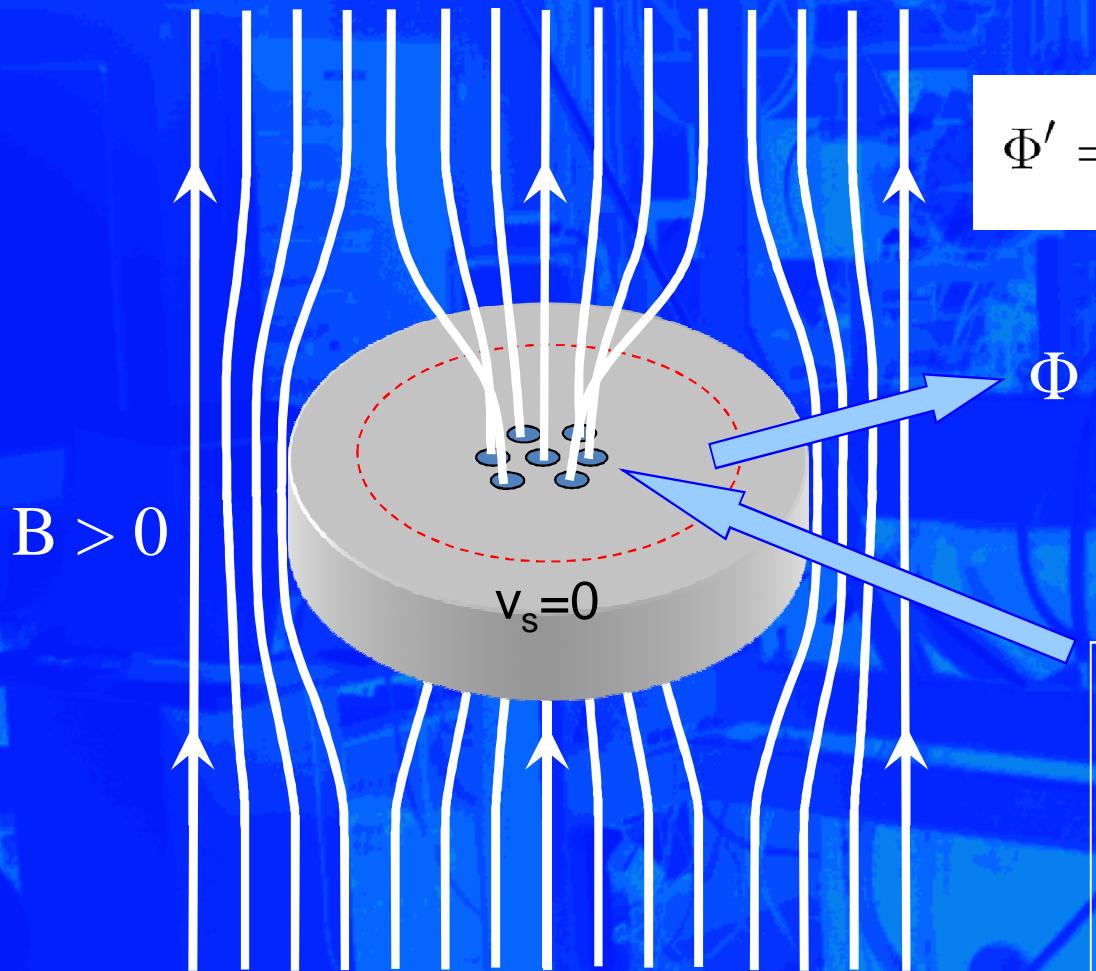
Superconductivity: Ginzburg-Landau Approach



Superconductivity: Ginzburg-Landau Approach



Superconductivity: Ginzburg-Landau Approach

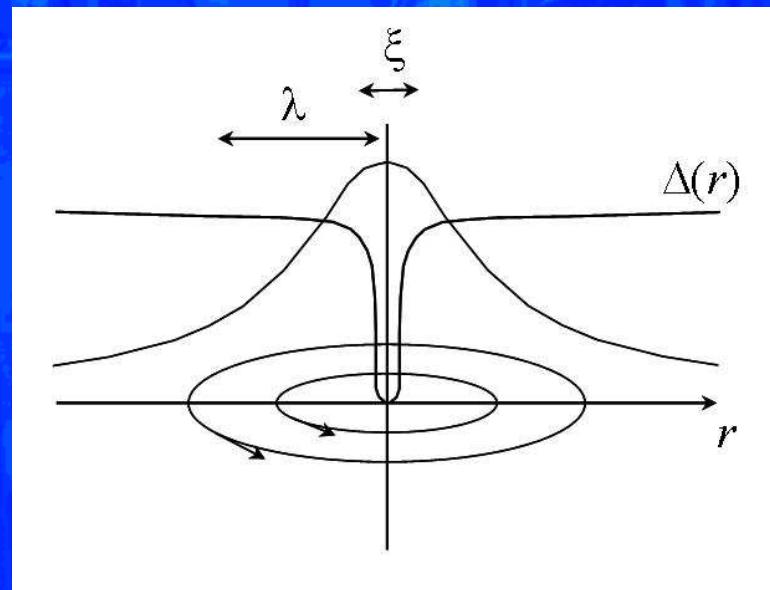


$$\Phi' = \Phi + \frac{1}{2e} \oint m^* \vec{v}_s \cdot d\vec{s} = n \frac{\hbar}{2e} = n\Phi_0$$

In type II superconductors ($k>1$) the Abrikosov vortex lattice forms, each vortex containing the flux quantum Φ_0

Superconductivity: Ginzburg-Landau Approach

Vortex Matter in Superconductors
(very huge field of research!)

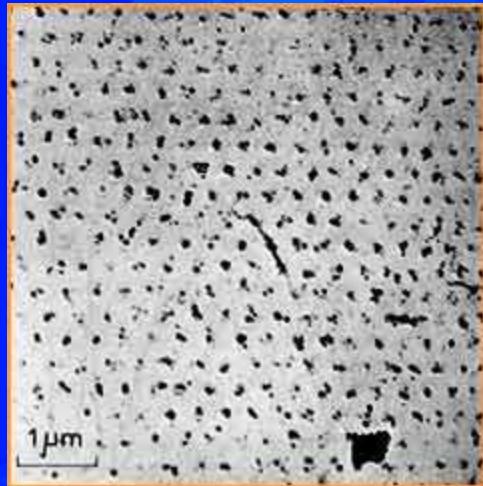


- N.B.1 : At the distance r , $|\vec{\nabla}\varphi| = 1/r$ → it diverges at $r=0$ ($v_s \rightarrow \infty$)
- N.B.2 : Flux quantification is valid over the area $S >> \pi\lambda^2$

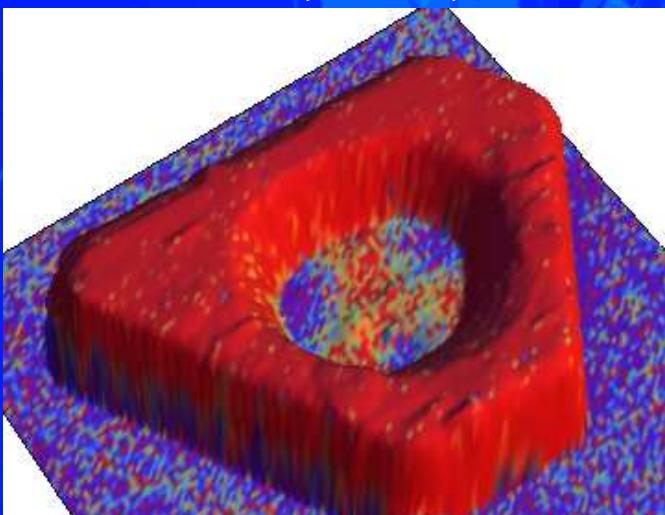
Vortex: General Property of Rotating Quantum Condensates

Superconductors (BCS)

First image of Vortex, 1967

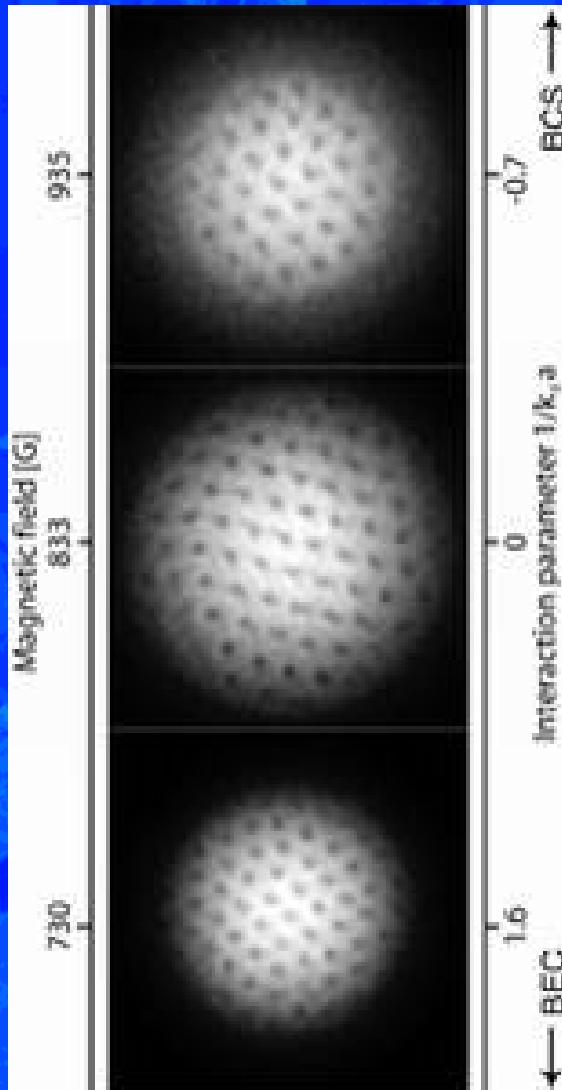


*3 vortices in SC nano-island
STM/STS, INSP, 2009*



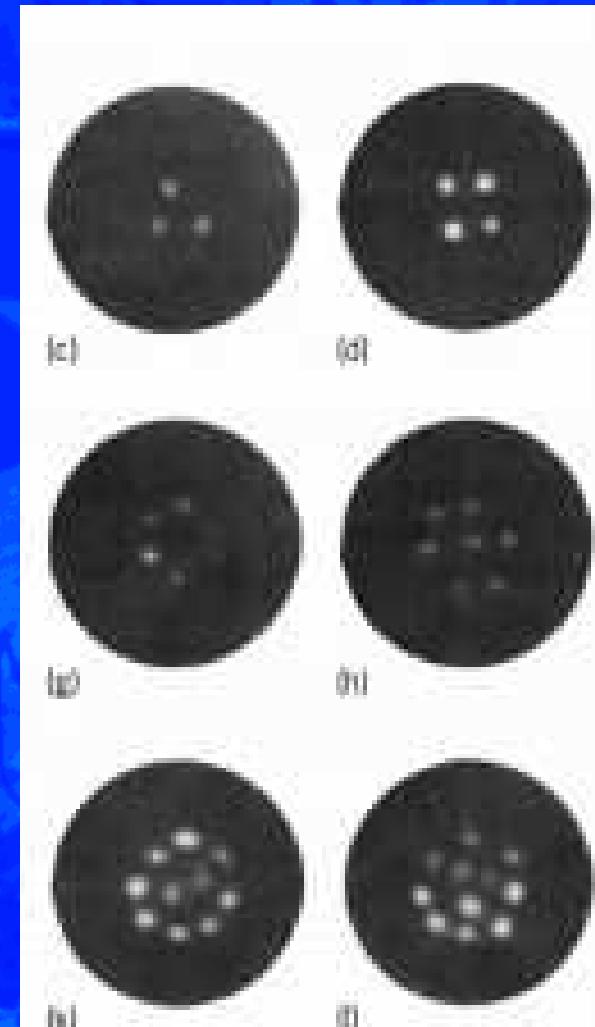
Cold atoms (BEC)

Vortex in ultracold condensate of atoms



Quantum liquids

Vortex in superfluid He



S



Tunneling into a Superconductor

$$\frac{dI}{dV}(V) = -\frac{2\pi e^2 T}{\hbar} N_p \int_{-\infty}^{\infty} N_s(E+eV) \frac{\partial f}{\partial E}(E) dE$$

BCS-kind Superconductor:
Gap 2Δ in the excitation spectrum:

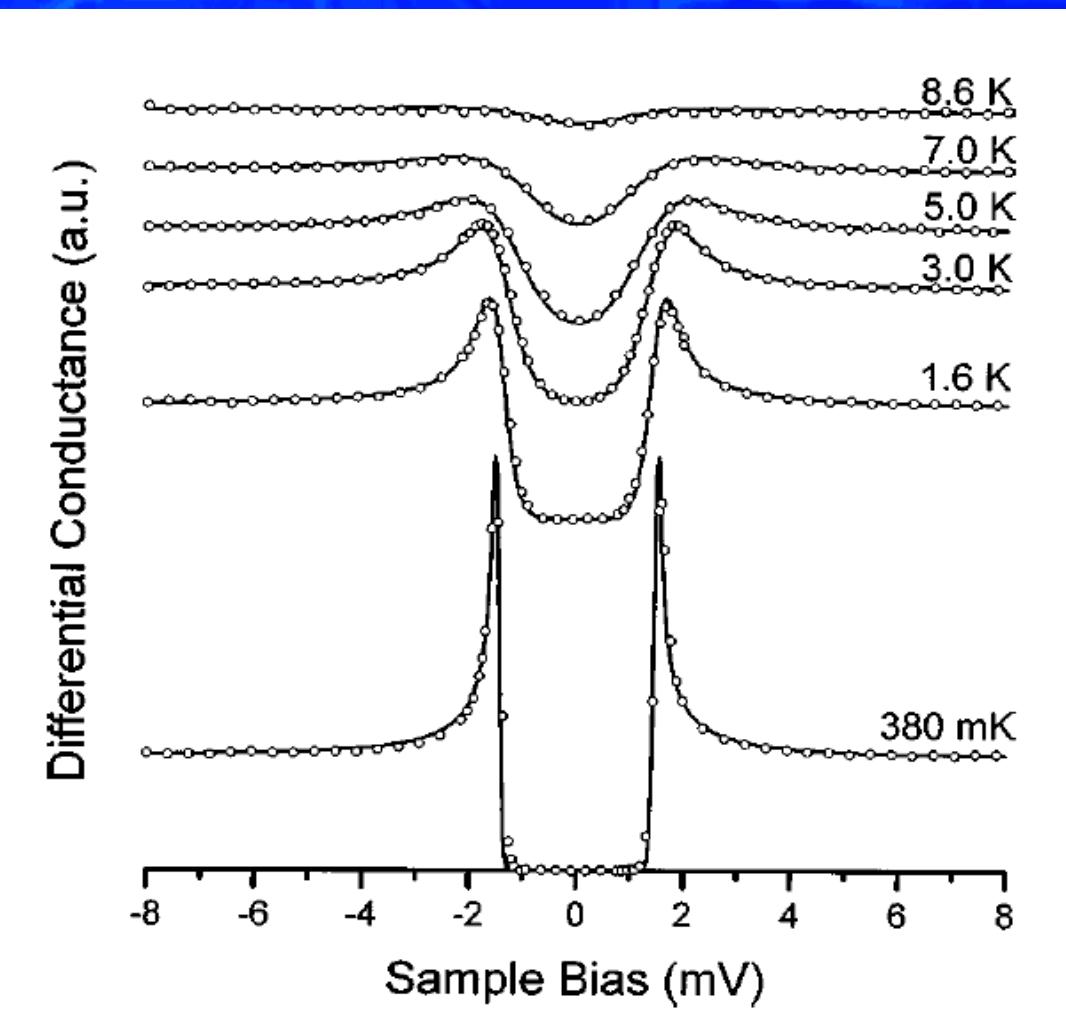
$$N_s(E) = N(0) \frac{E}{\sqrt{(E^2 - \Delta^2)}}$$



BCS

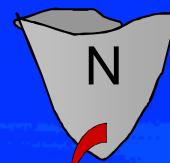


$$\frac{2 \Delta(0)}{k T_C} \approx 3.5$$

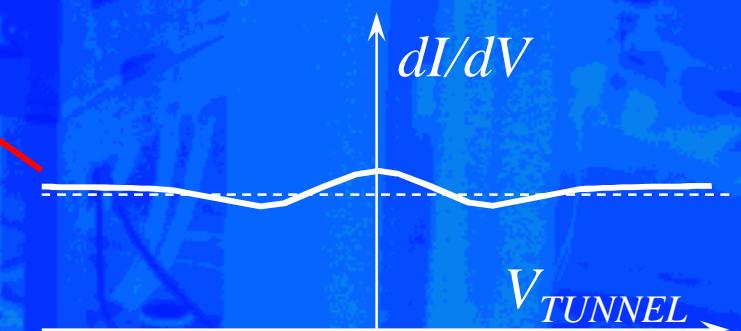
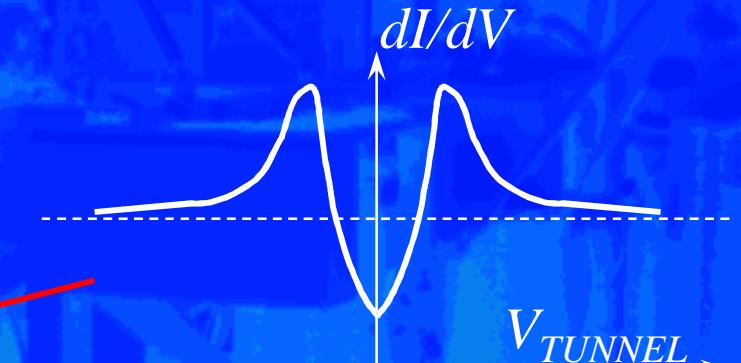
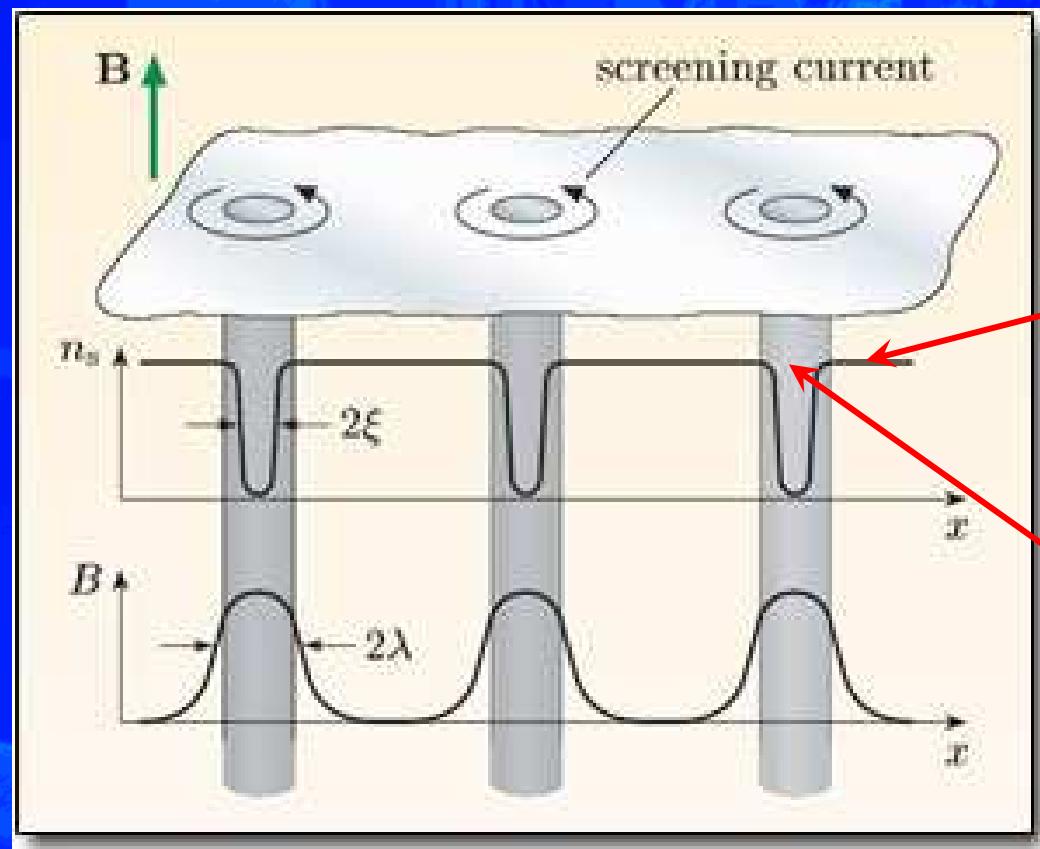


Pan, Davis, Univ. de Berkeley.(APL, 1998)

Ecole MICO 2010



Abrikosov Vortex Lattice observed by Scanning Tunneling Spectroscopy (STS)

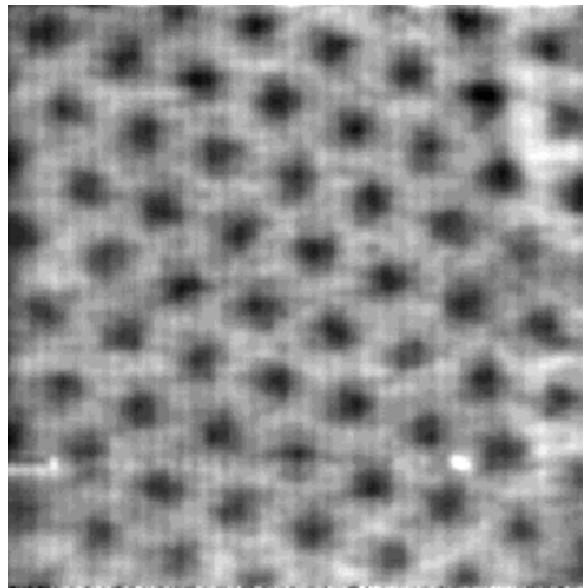


$$dI / dV(\mathbf{r}) \propto N(E, \mathbf{r})$$

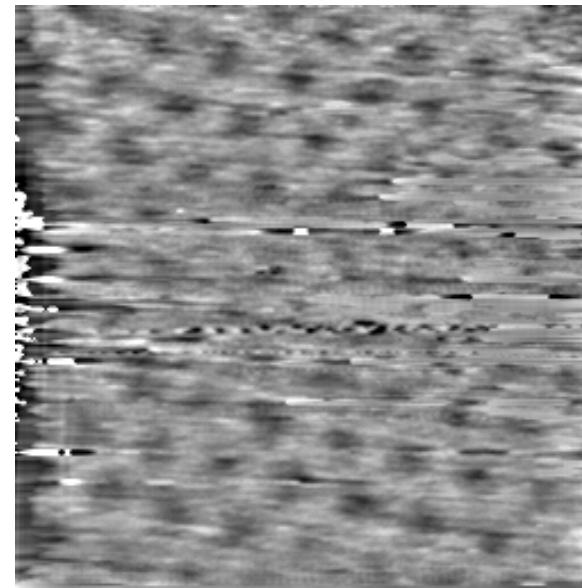
Vortex Lattice Study (still picture)

Vortex lattice prepared in 2H-NbSe₂ at a given magnetic field:

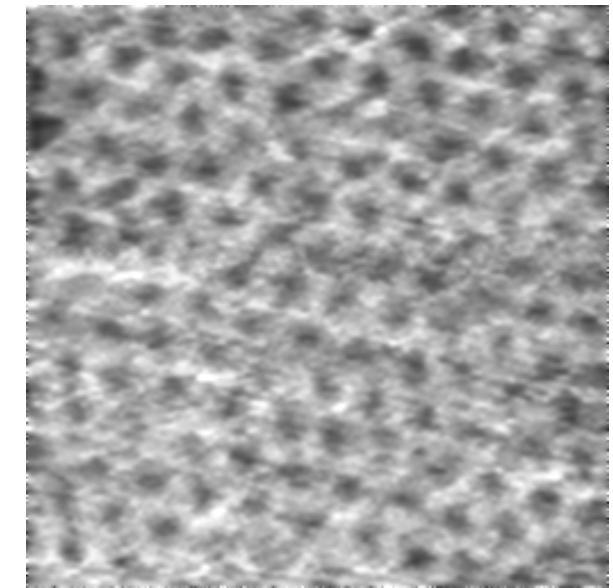
0.5T



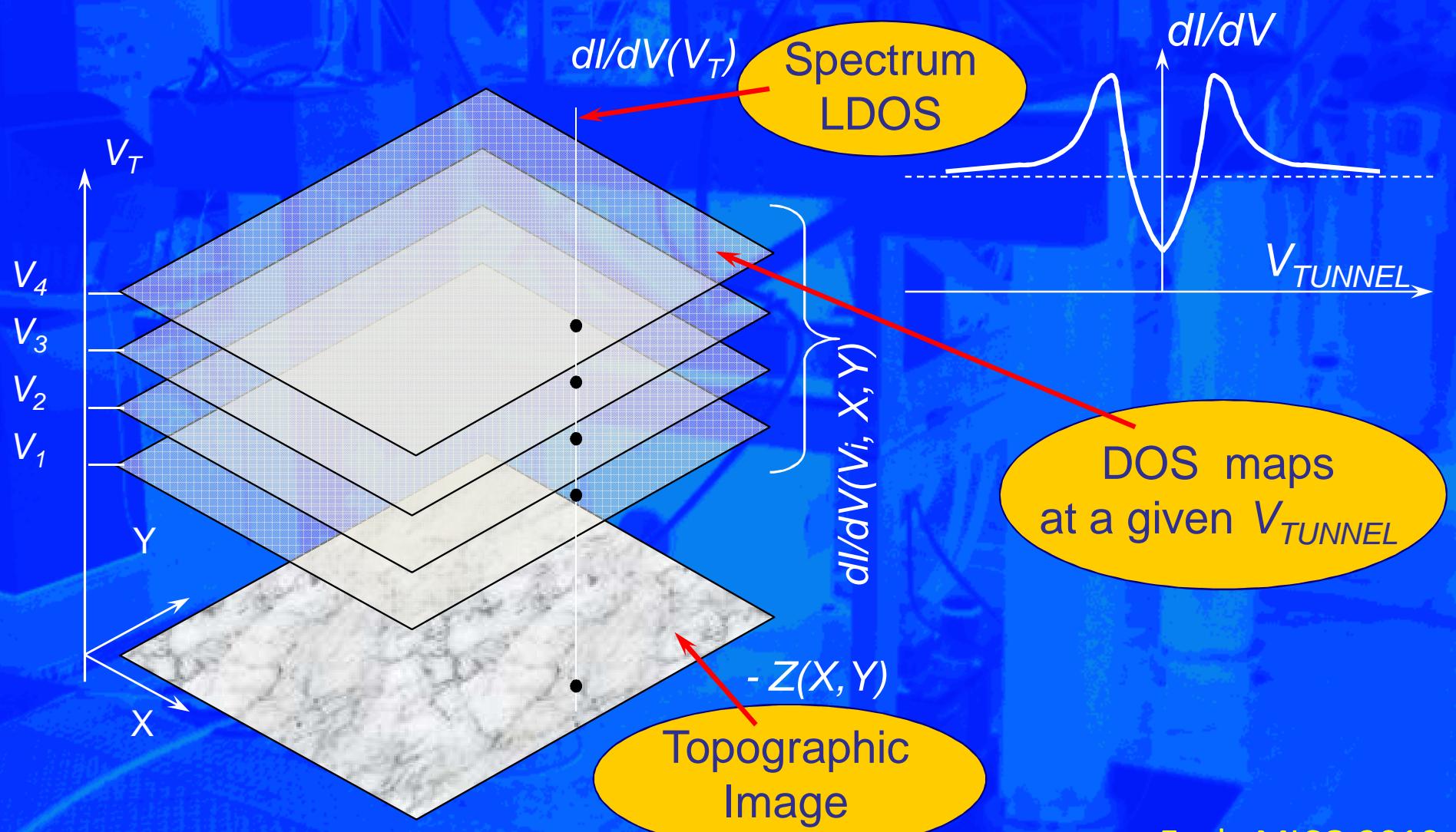
0.8T



1.7T



STM/STS : Scanning Tunneling Spectroscopy



OUTLINE

Scanning Tunneling Microscopy (STM) and Spectroscopy (STS)

1. Physical background
2. How to make it, how it looks like
3. Spectroscopy of Superconductors → MgB_2
4. Other strongly correlated materials

S



Tunneling into a Superconductor

$$\frac{dI}{dV}(V) = -\frac{2\pi e^2 T}{\hbar} N_p \int_{-\infty}^{\infty} N_s(E+eV) \frac{\partial f}{\partial E}(E) dE$$

BCS-kind Superconductor:
Gap 2Δ in the excitation spectrum:

$$N_s(E) = N(0) \frac{E}{\sqrt{(E^2 - \Delta^2)}}$$

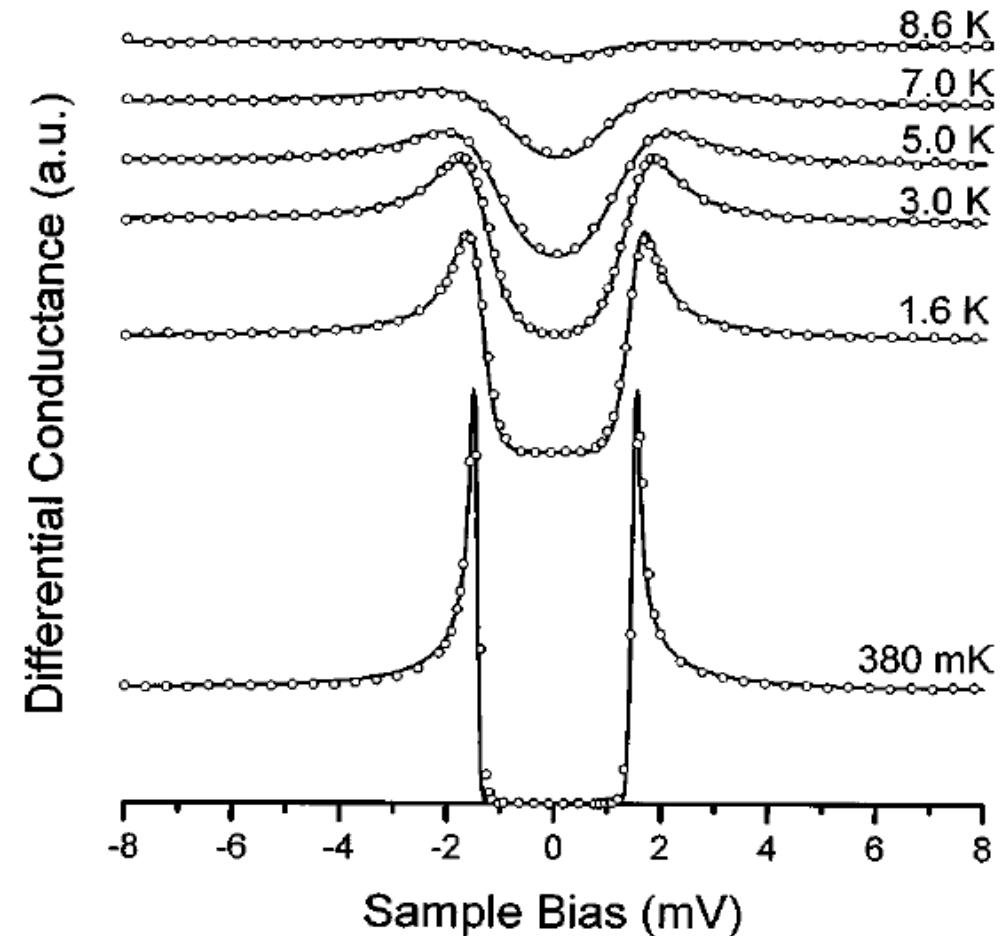


BCS



$$\frac{2 \Delta(0)}{k T_c} \approx 3.5$$

MgB_2 : $T_c=39\text{K}$ $\rightarrow \Delta(0)=6\text{mV}$



Pan, Davis, Univ. de Berkeley. (APL, 1998)

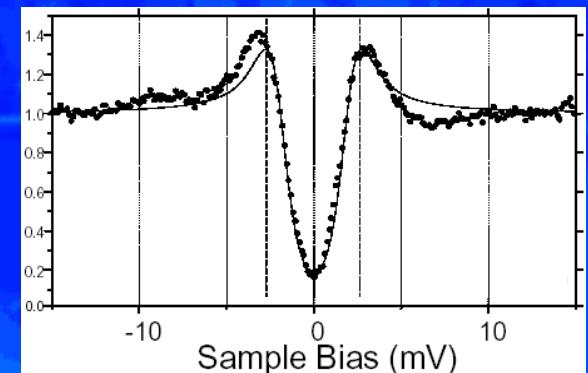
STS on Granular MgB₂

Result on more than 10^6 tunneling spectra locally measured:

- On ~99% of spectra : Single Gap DOS:

Small Gap Energy : $\Delta = 2.5 \pm 0.8$ meV

Strong « Pair-Breaking » : $\Gamma = 0.3-0.5$ meV



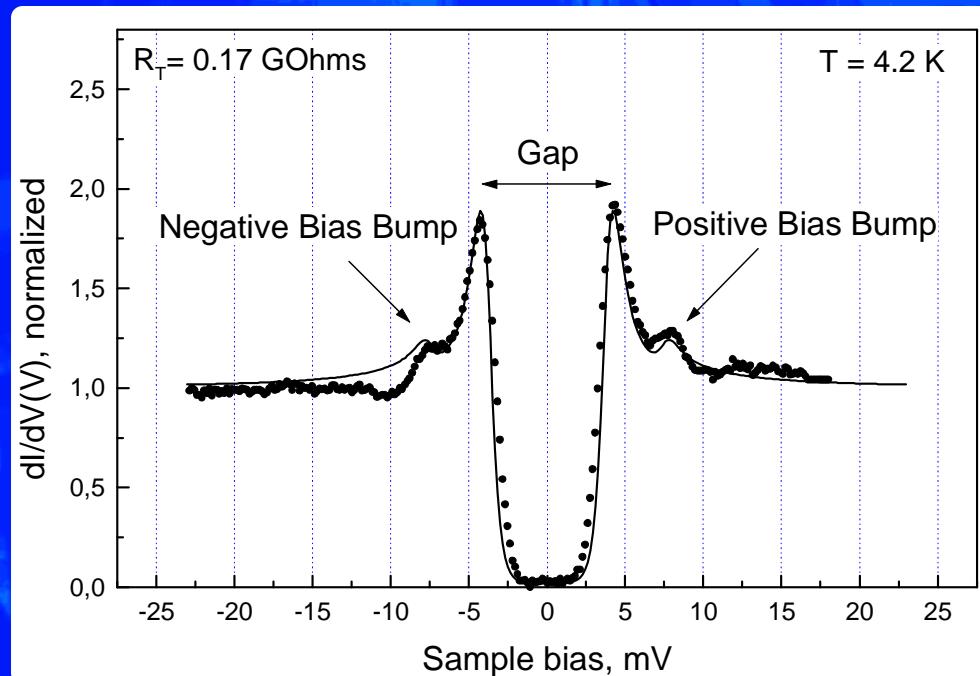
- On ~1% of spectra (hundreds spectra): Some « strange » DOS!



???

STS on Granular MgB₂

Fit : weighted sum of two BCS DOS



$$\rho_S^L = \Re e \left[\frac{\varepsilon - i\Gamma}{\sqrt{(\varepsilon - i\Gamma)^2 - \Delta_L^2}} \right]$$

$$\rho_S^S = \Re e \left[\frac{\varepsilon - i\Gamma}{\sqrt{(\varepsilon - i\Gamma)^2 - \Delta_S^2}} \right]$$

$$\rho_N(\varepsilon) = \text{Const}$$

$\sigma^{LS} \propto \int_{-\infty}^{+\infty} g(\varepsilon, V) \rho_S^{LS}(\varepsilon - eV) d\varepsilon$

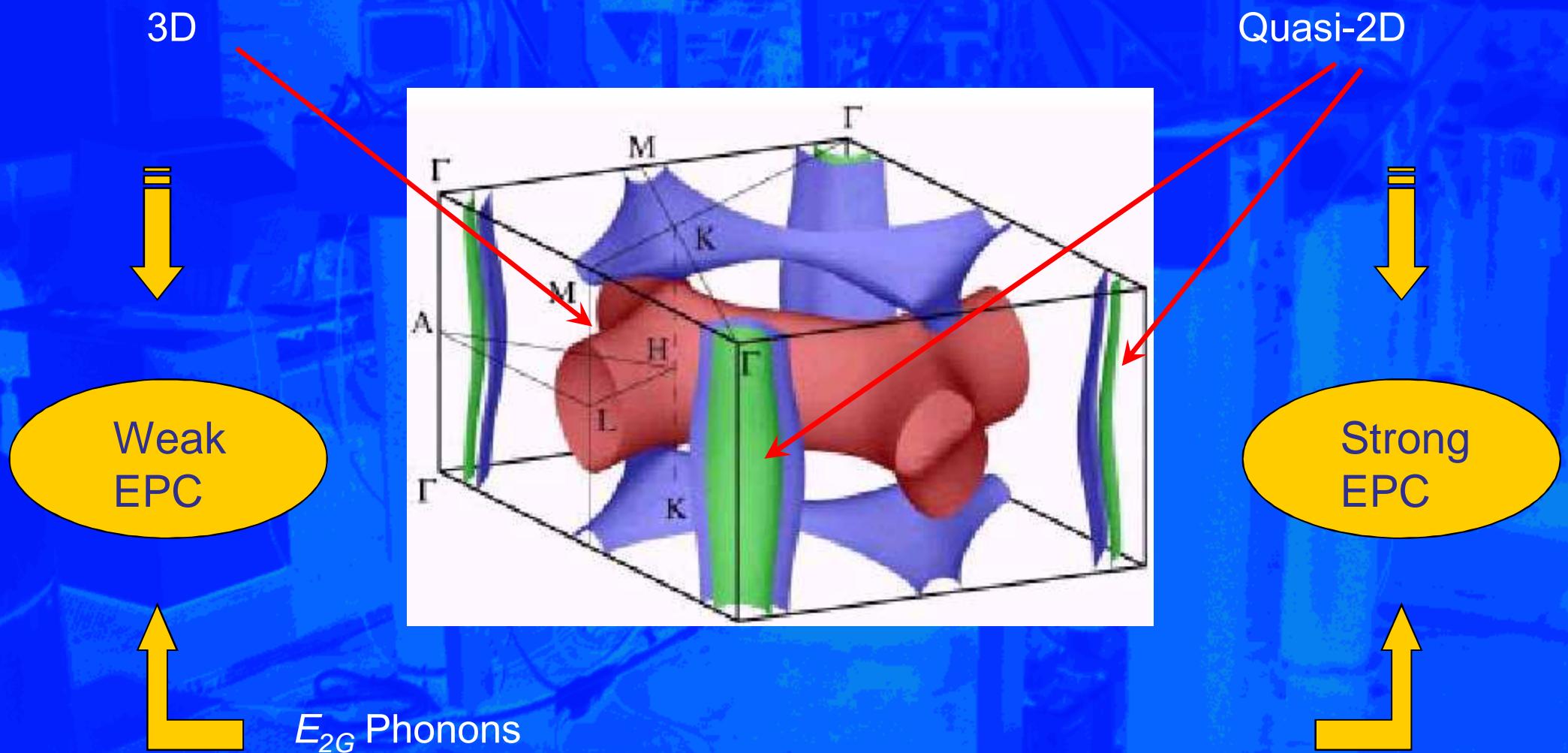
$\sigma \propto C \times \sigma^L + (1-C) \times \sigma^S$

$C = 0.13 \rightarrow \Delta_L = 7.4$ mV
 $1-C = 0.87 \rightarrow \Delta_S = 3.5$ mV
 $\Gamma = 0.15$ mV

STS on Granular MgB₂ : Questions

- ➡ « Strange » spectra are rare— are they relevant?
- ➡ Why such a statistic (1%)?
- ➡ What is the physical background of the effect?

MgB₂ : Electronic Structure

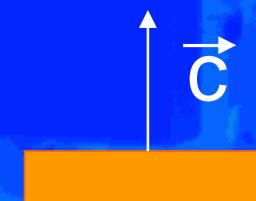


STS on Granular MgB₂

→ MgB₂ is anisotropic material



The form of pellets:



→ Complex Fermi surface : 2D (ab-plane) and 3D

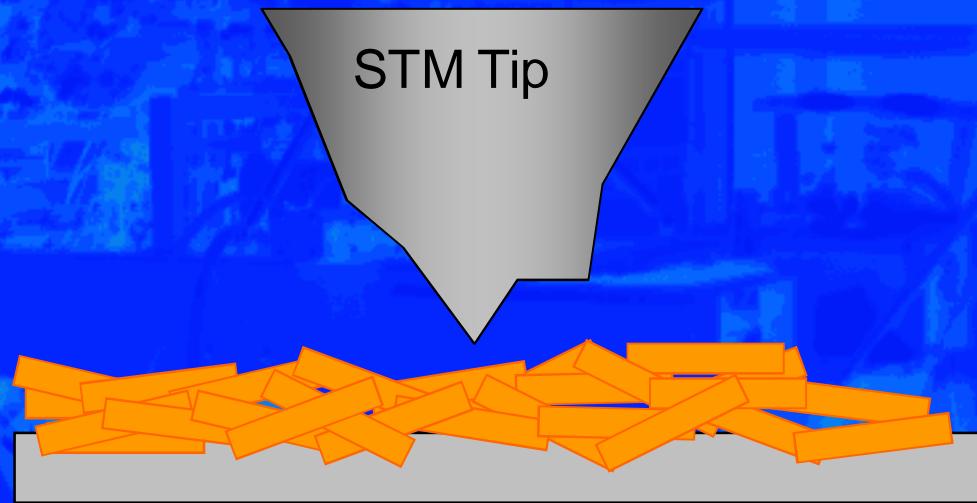


Orientation-dependent Tunneling!

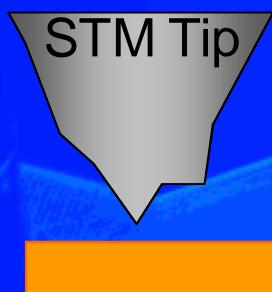
STS on Granular MgB₂



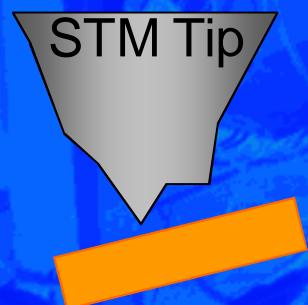
STM/STS on pellets:



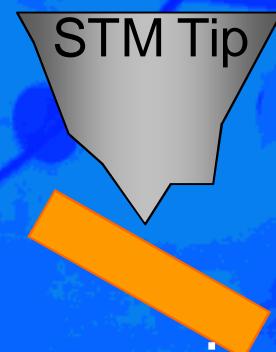
Local Geometry favours the tunneling into the 3D-band



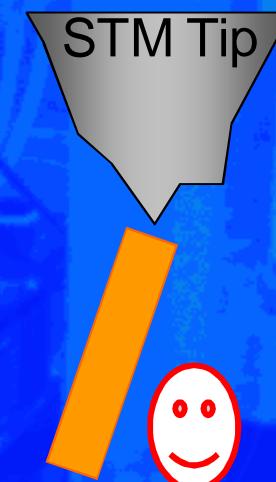
c-axis



c-axis

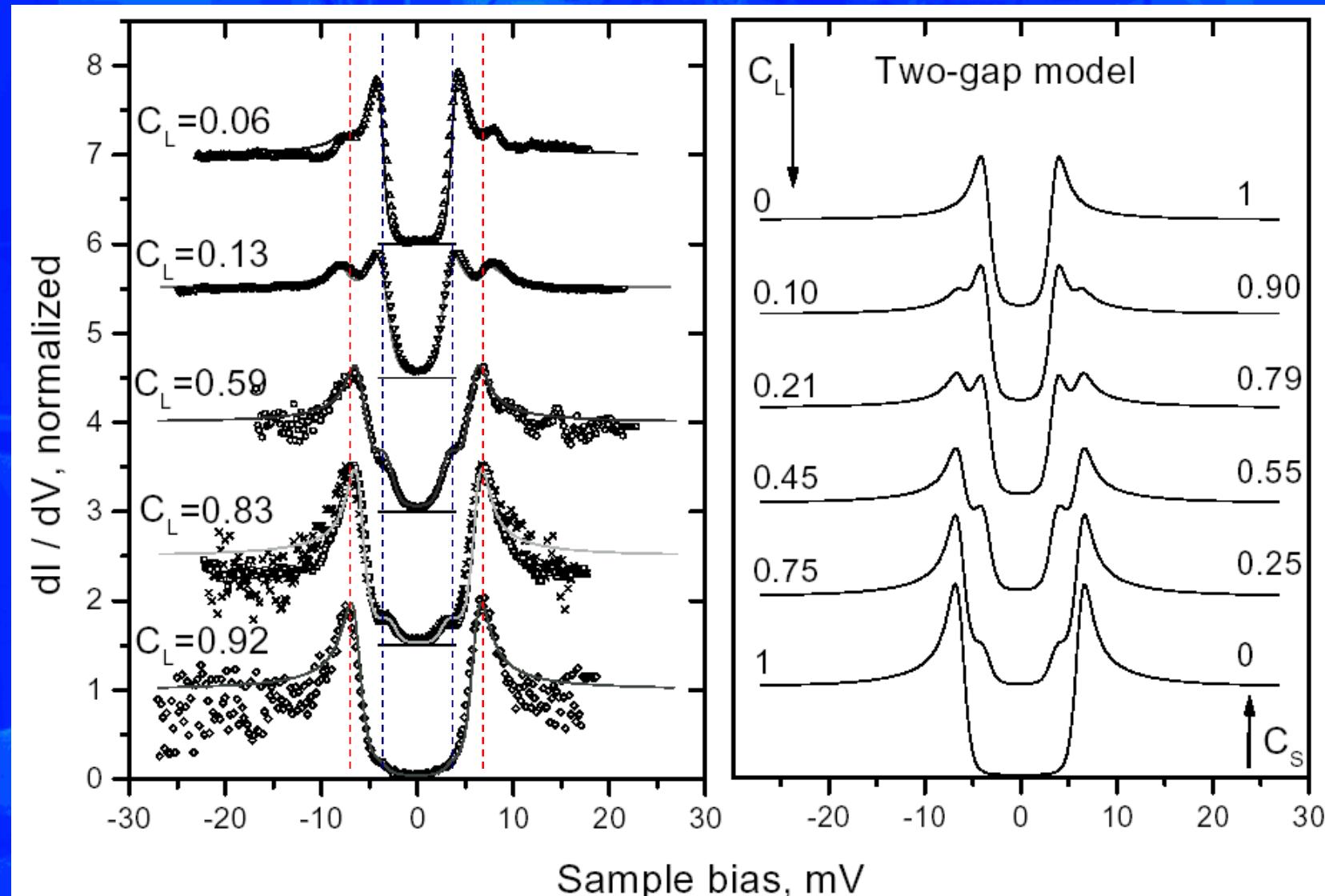


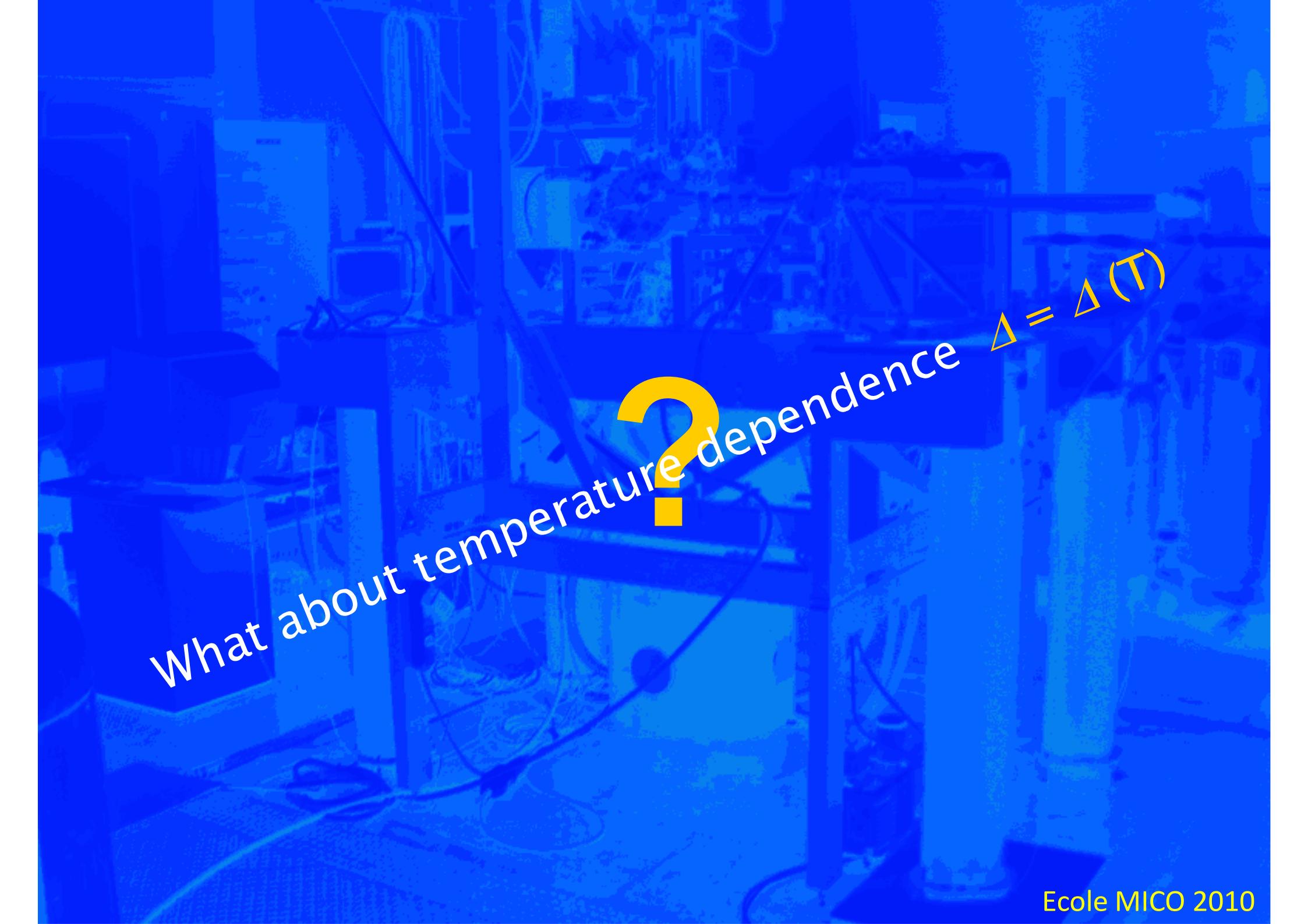
c-axis



STS on Granular MgB₂

- Selected : SIN spectra with different 2D/3D tunneling weights

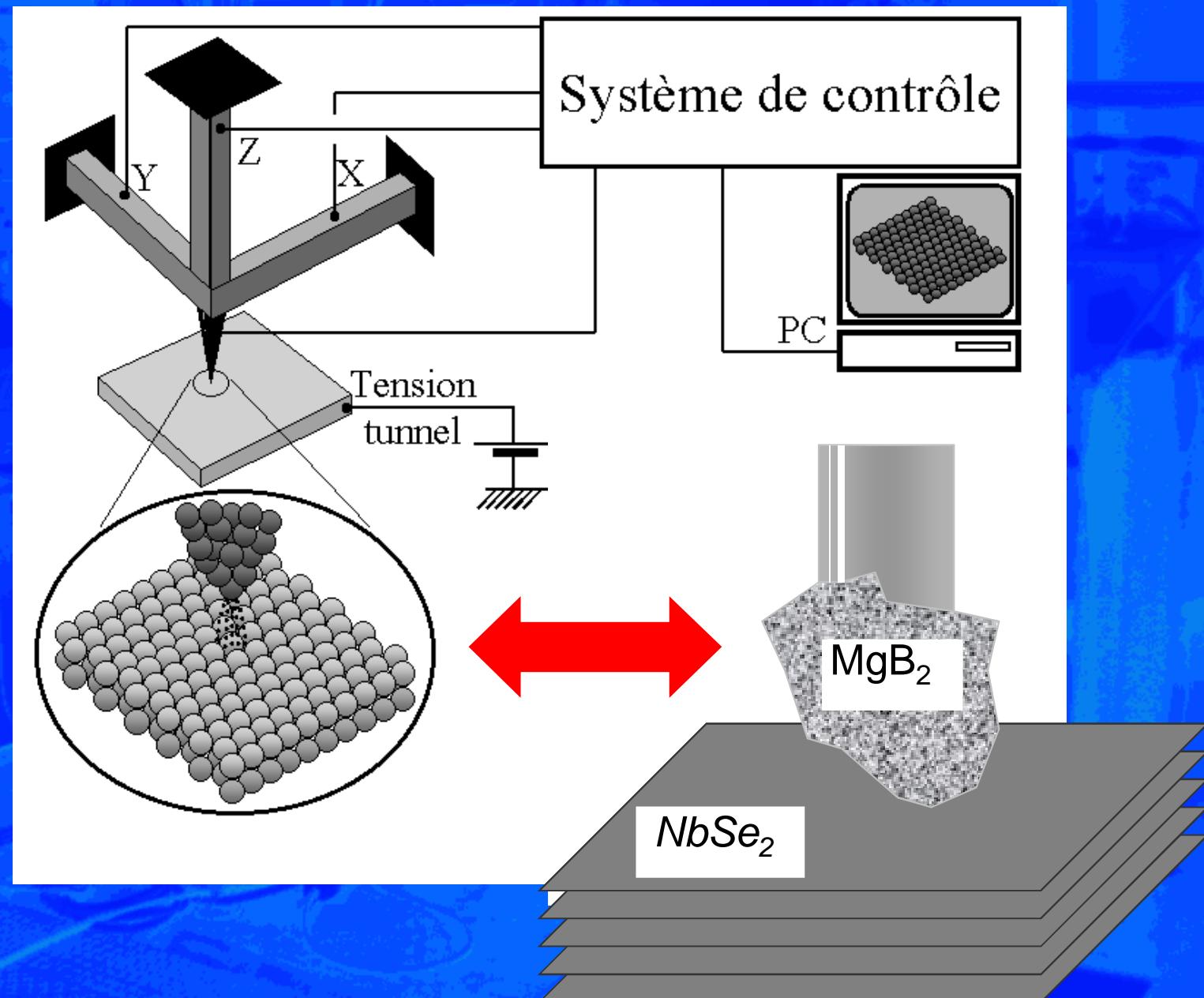




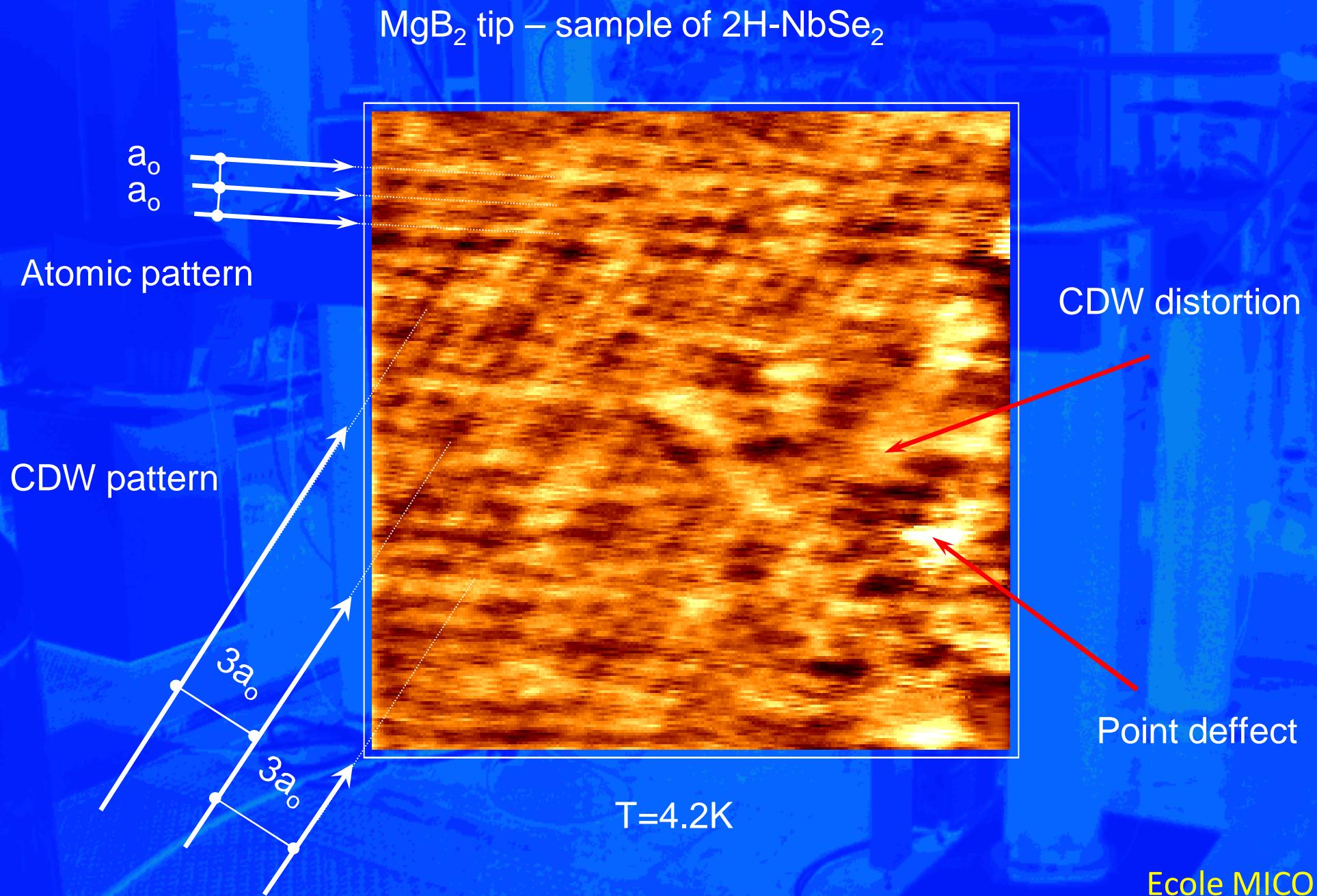
What about temperature dependence $\Delta = \Delta(T)$

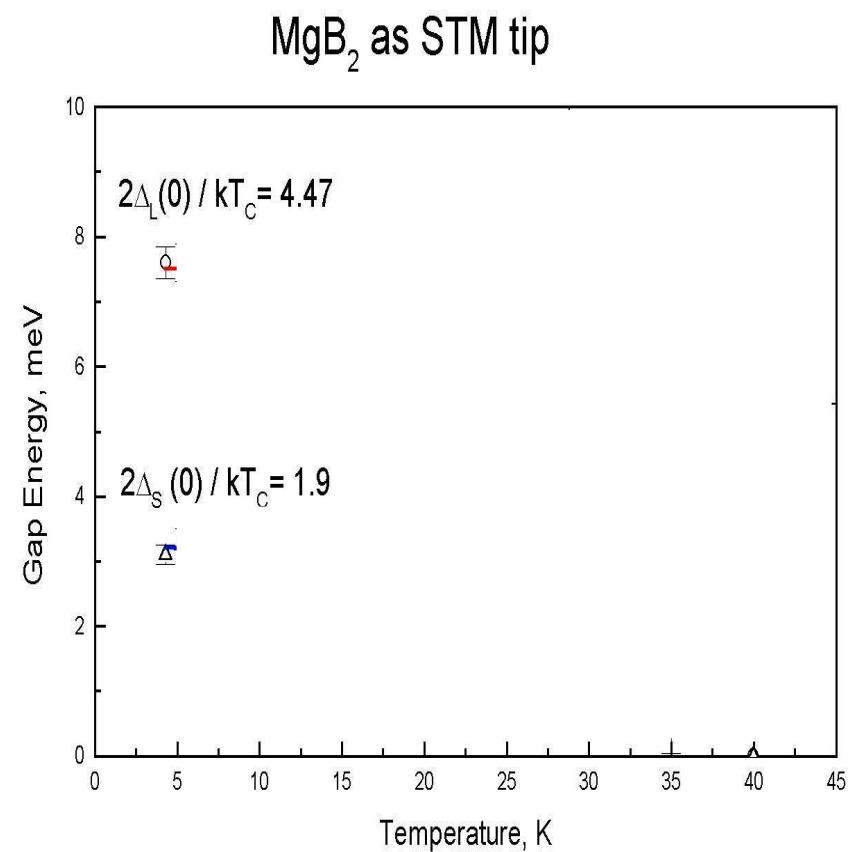
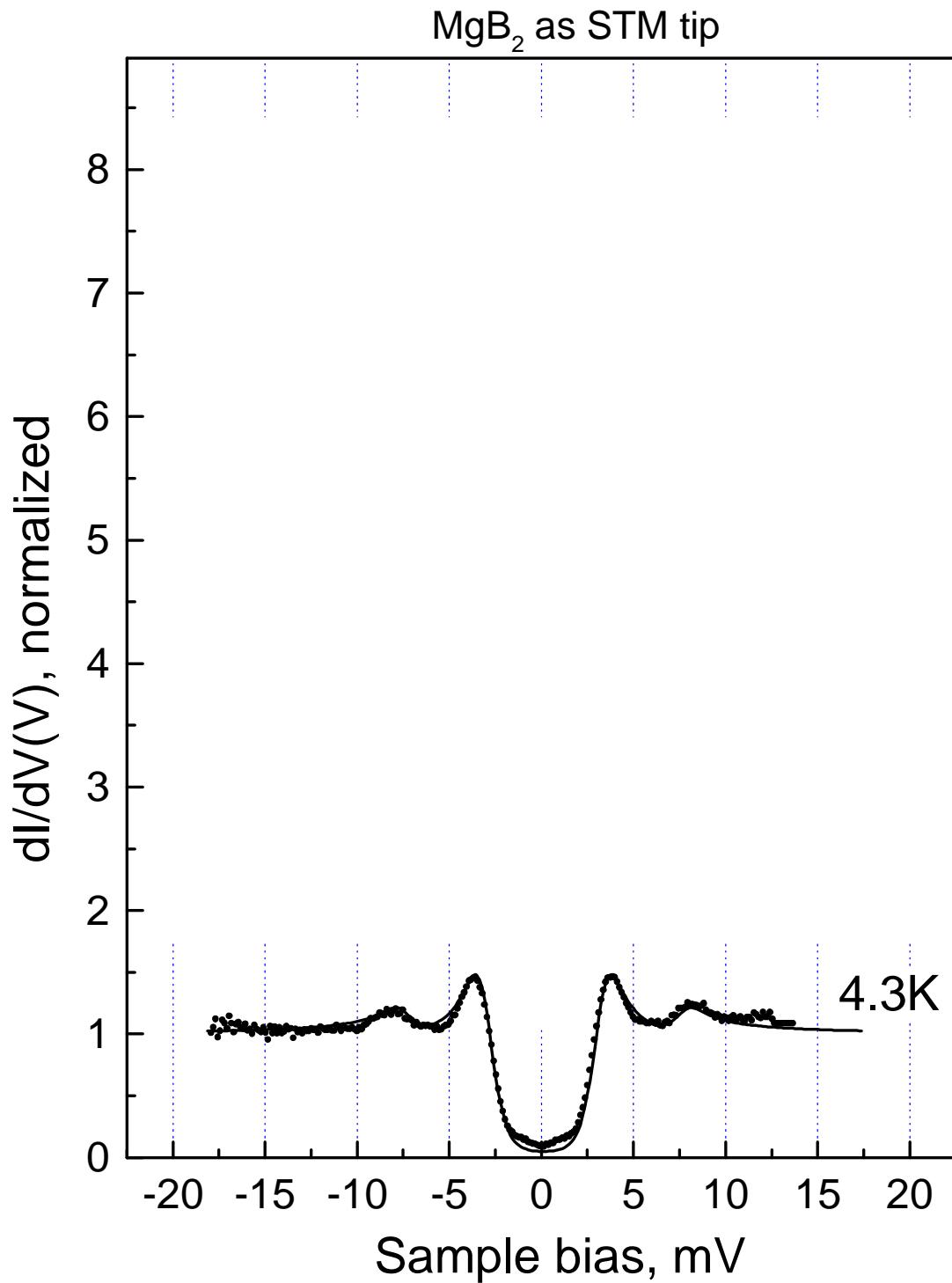
?

Inverted Junction Experiment



Inverted Junction Experiment



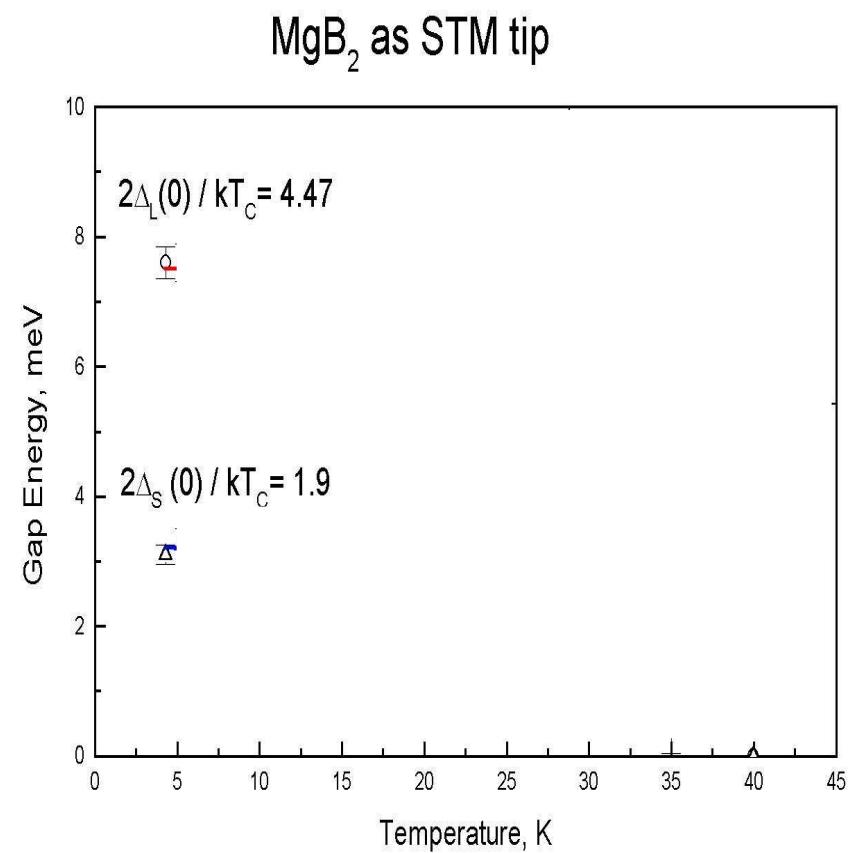
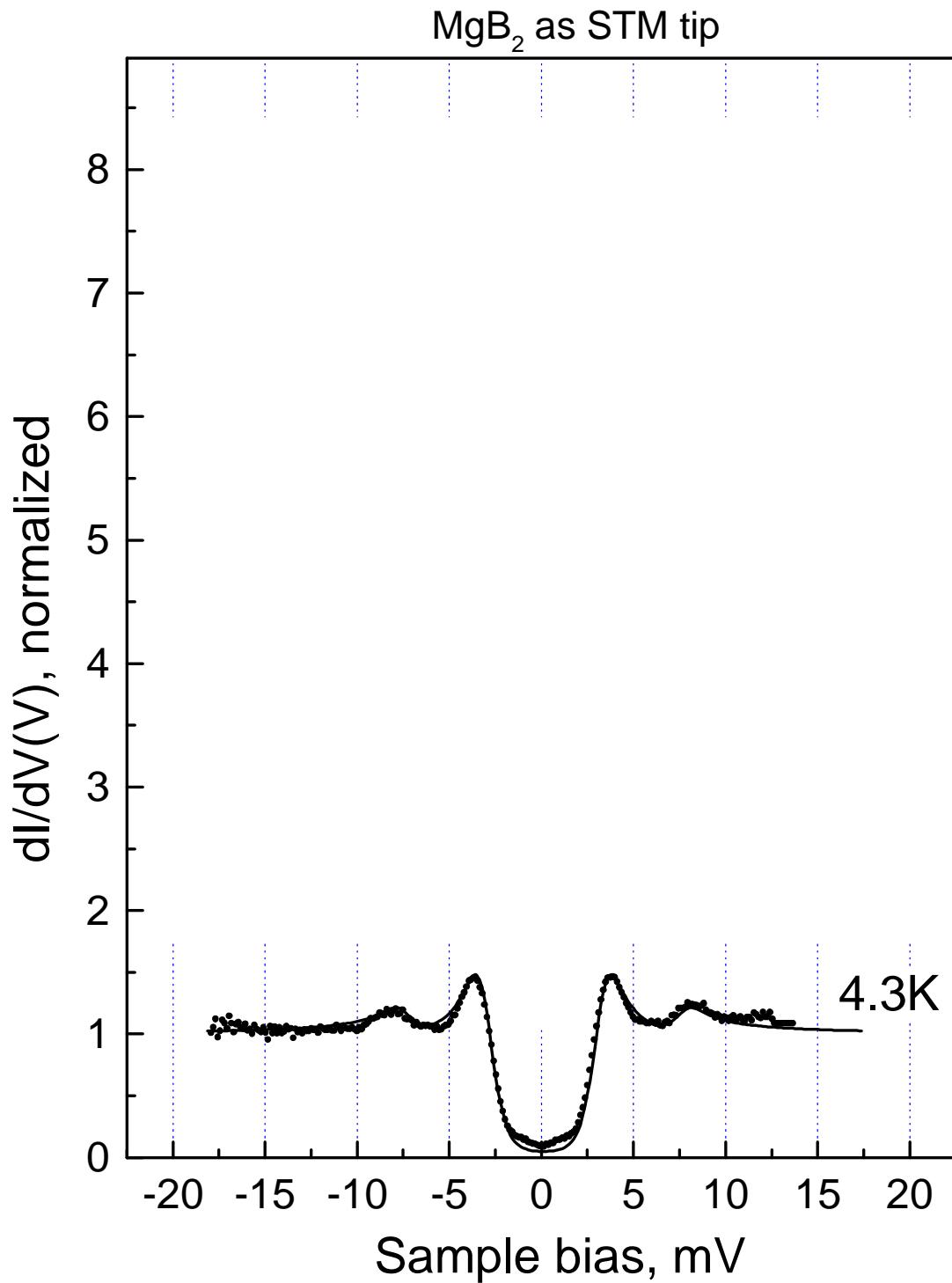


$$\sigma \propto C \times \sigma^L + (1-C) \times \sigma^S$$



Free Parameters :

$$\Delta_L, \Delta_S, C, \Gamma$$

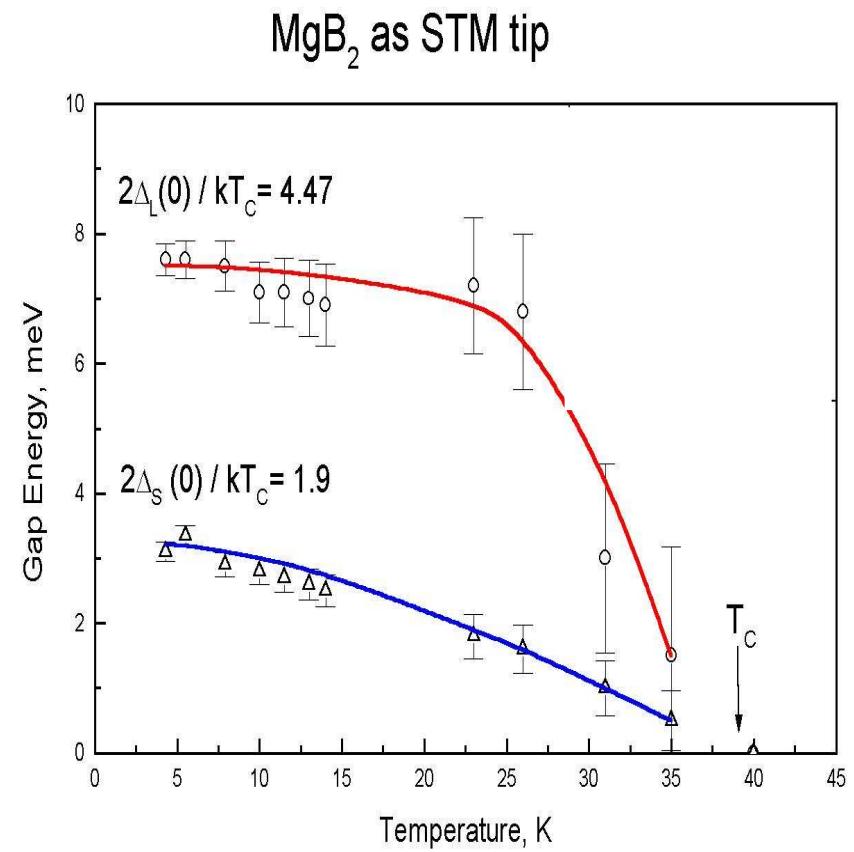
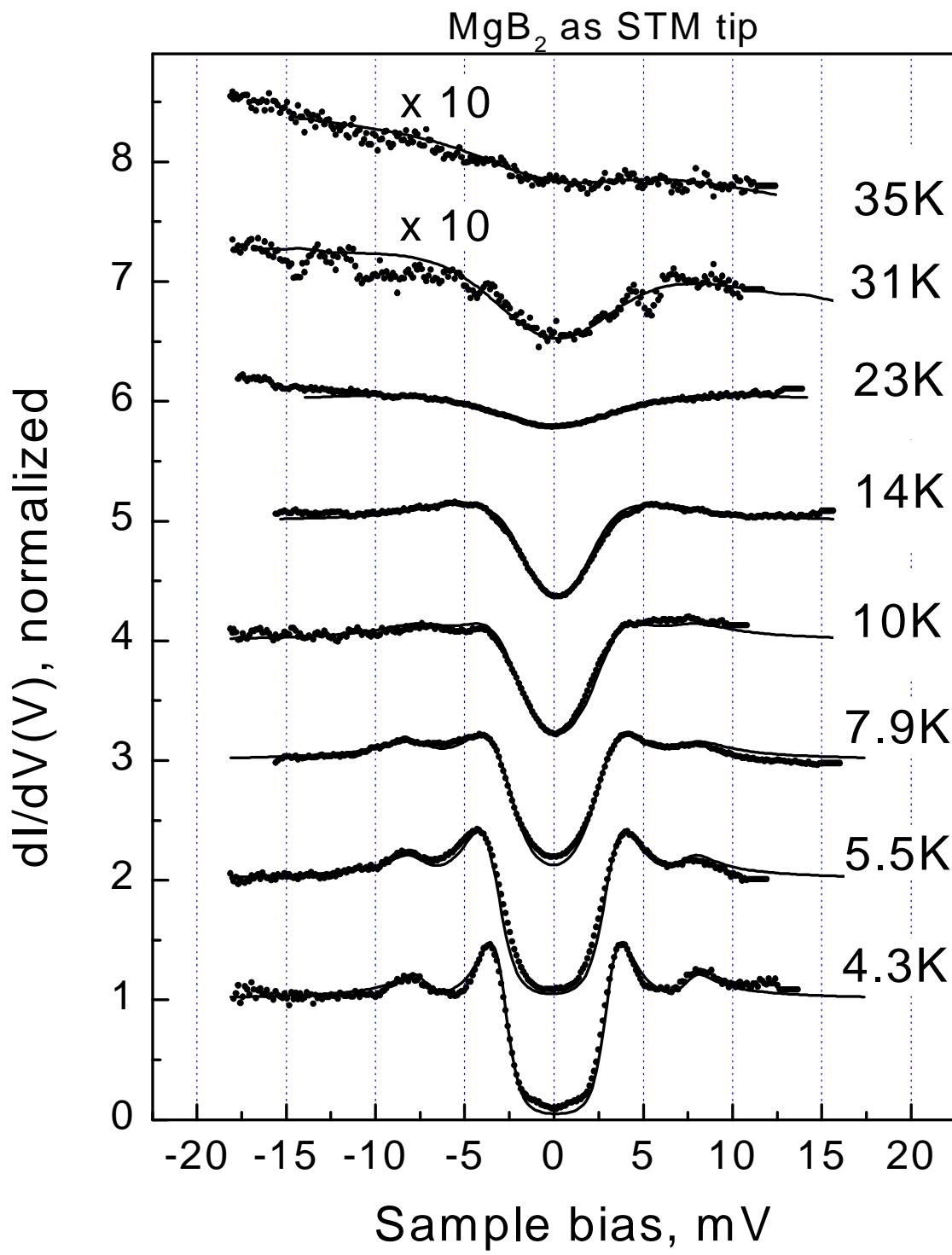


$$\sigma \propto C \times \sigma^L + (1-C) \times \sigma^S$$

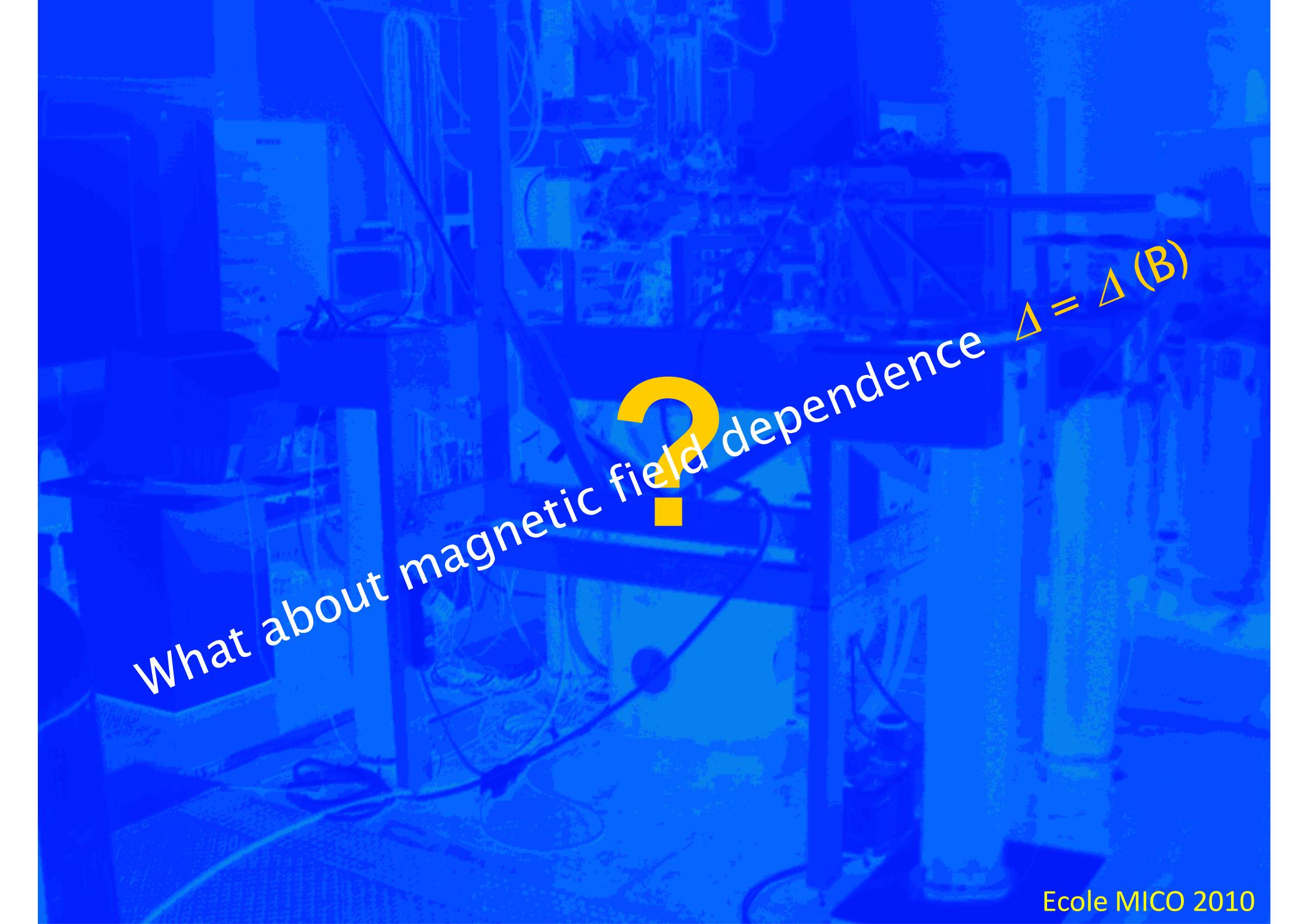


Free Parameters :

Δ_L, Δ_S, C, T

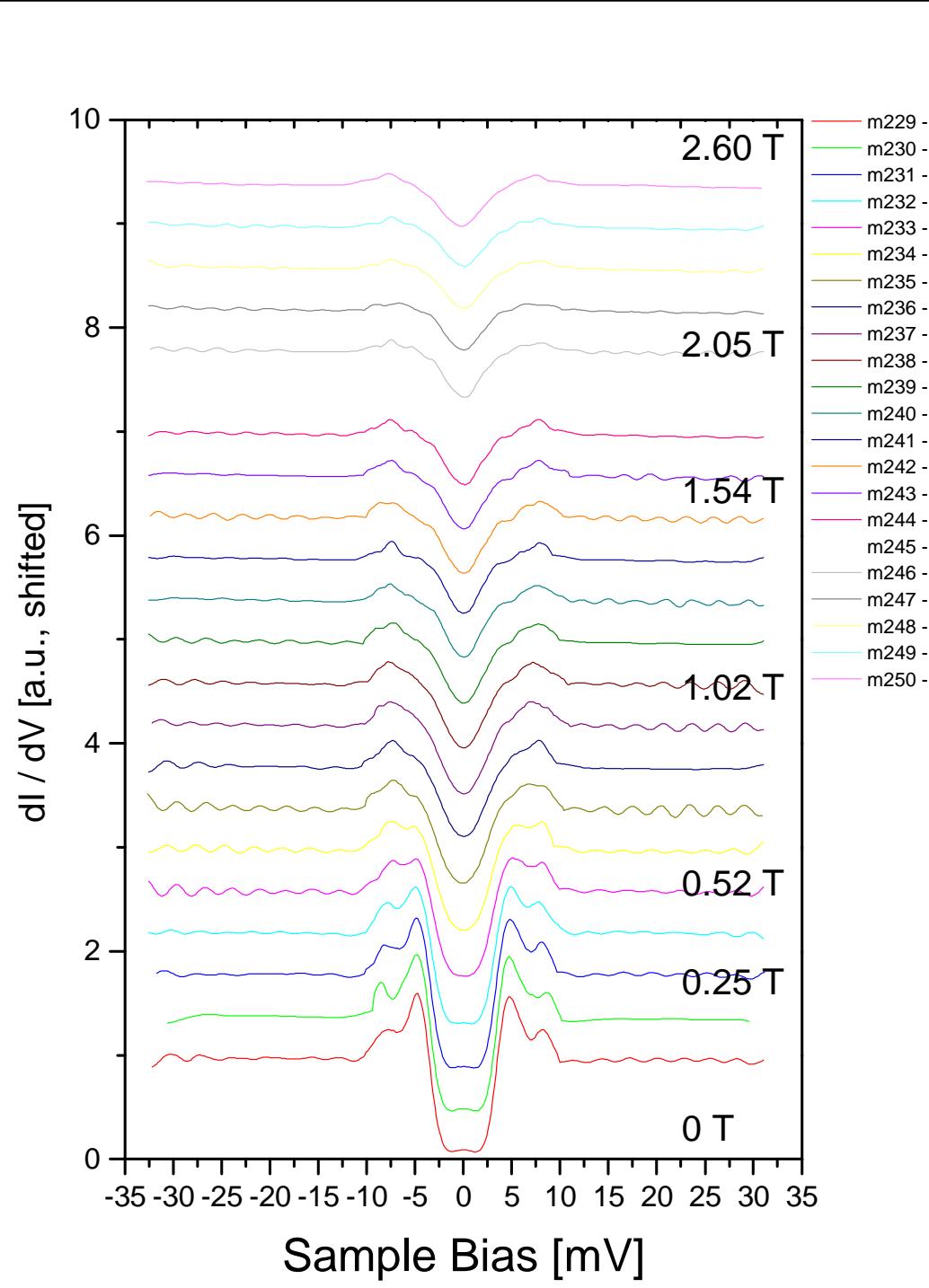


F. Giubileo et al.
cond-mat/0105592, Phys. Rev. Lett. 87
(2001)



What about magnetic field dependence $\Delta = \Delta(B)$

Magnetic Field Dependence



● Δ_S , Δ_S versus (B)

● Experiment:
- both « gaps » are present at 2T!
- large gap resists much better

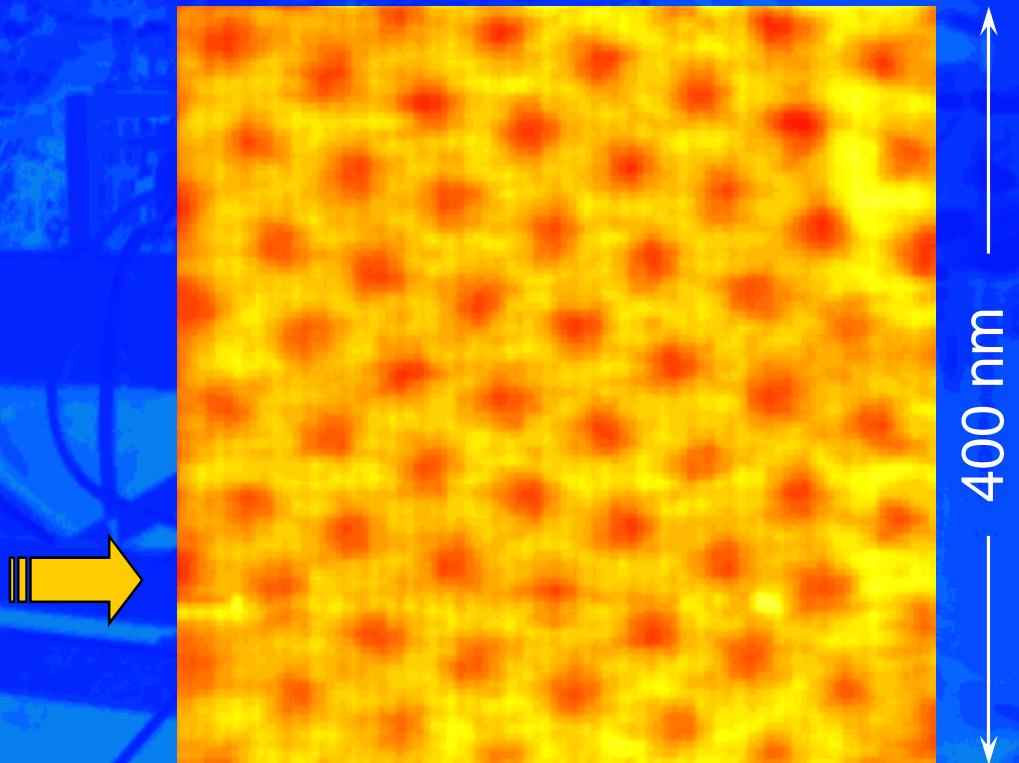
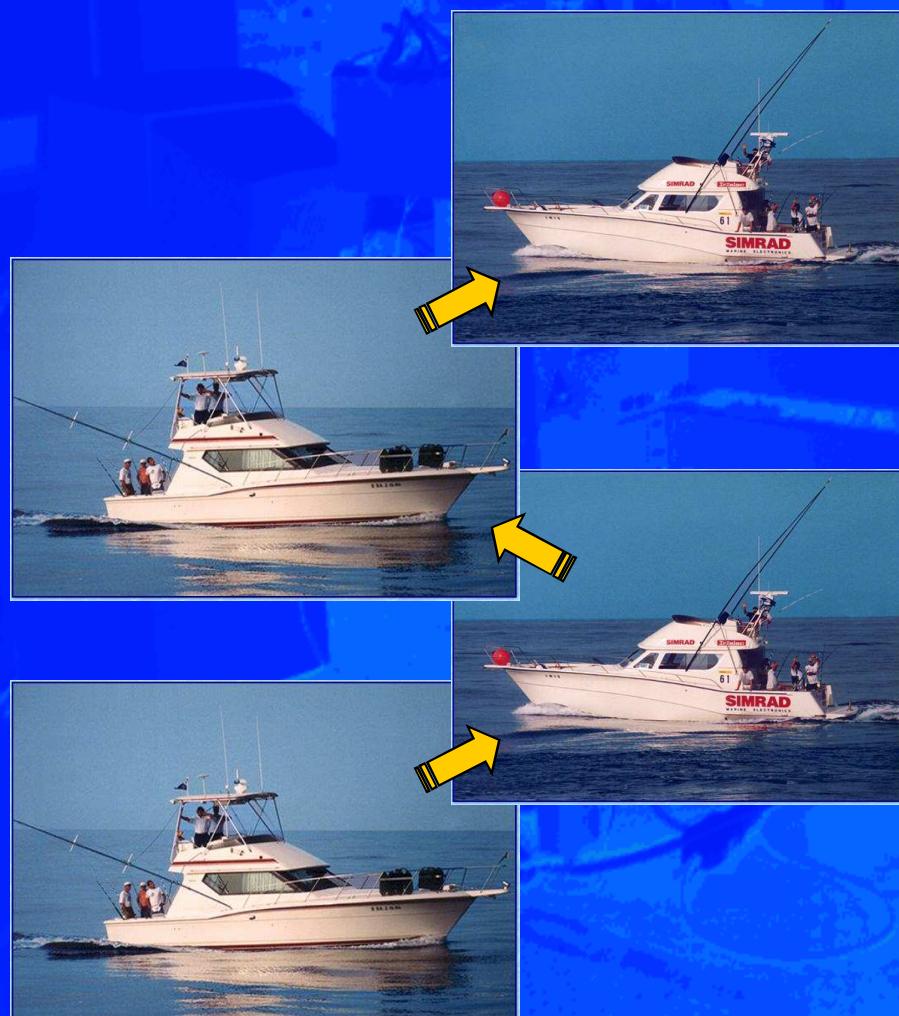
« Lazy Fisherman » Method of Vortex Study

Normal Fisherman



Fishing Vortices

Scanning Spectroscopy

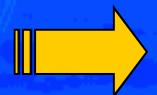


$T = 4.2 \text{ K}$

$B = 1.0 \text{ T}$

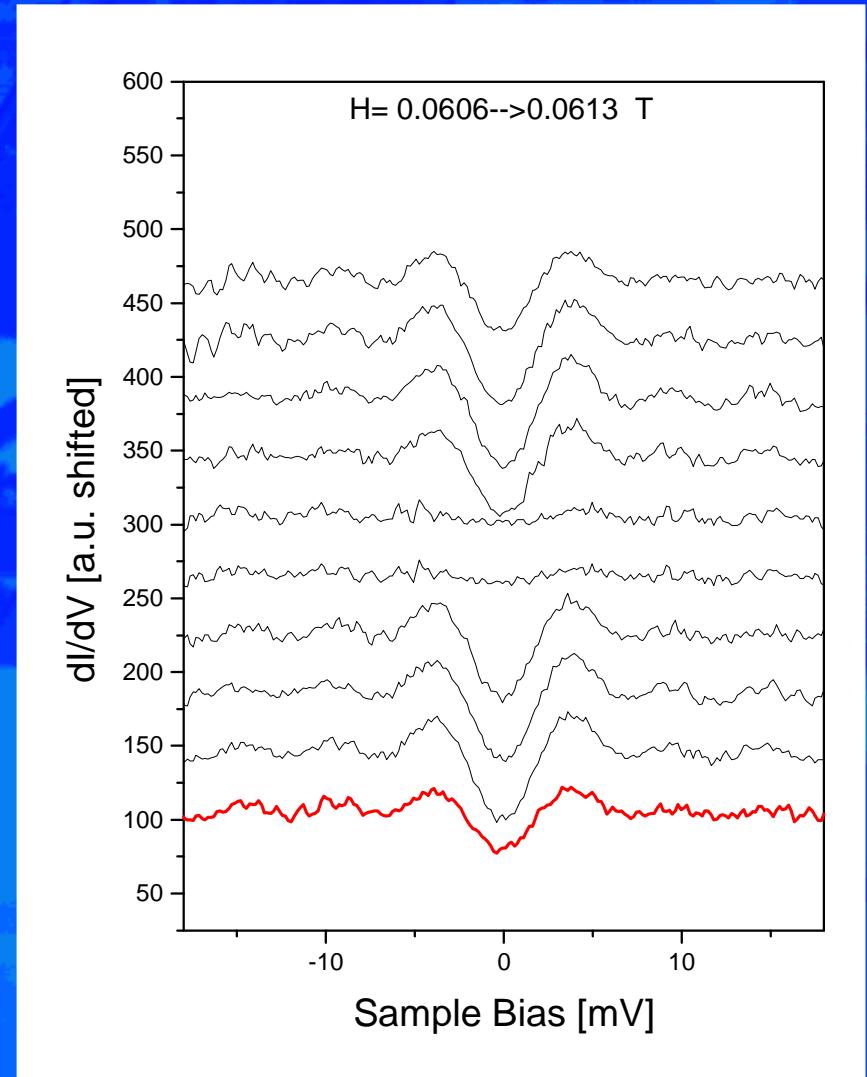
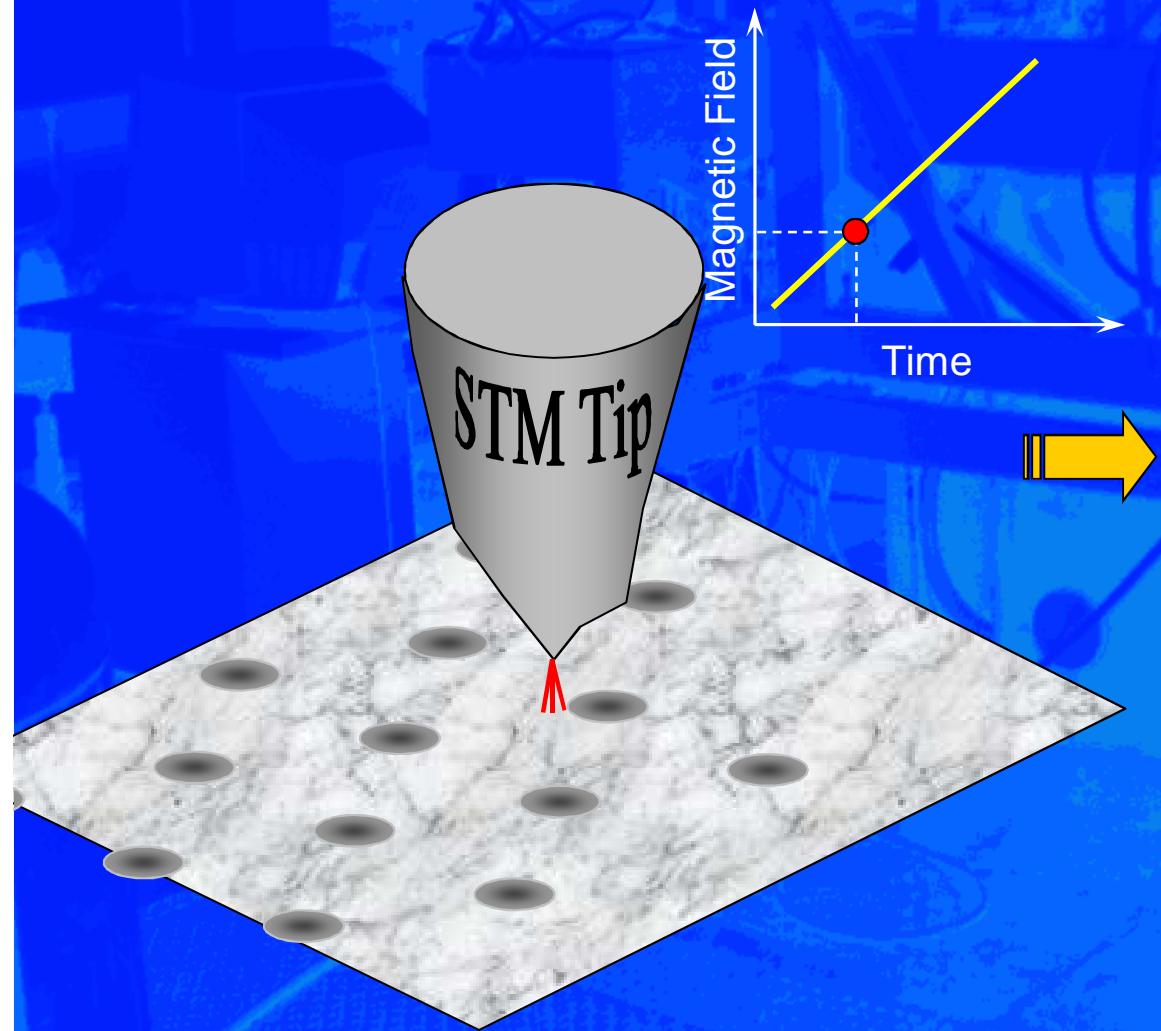
Fishing Vortices

Lazy Fisherman



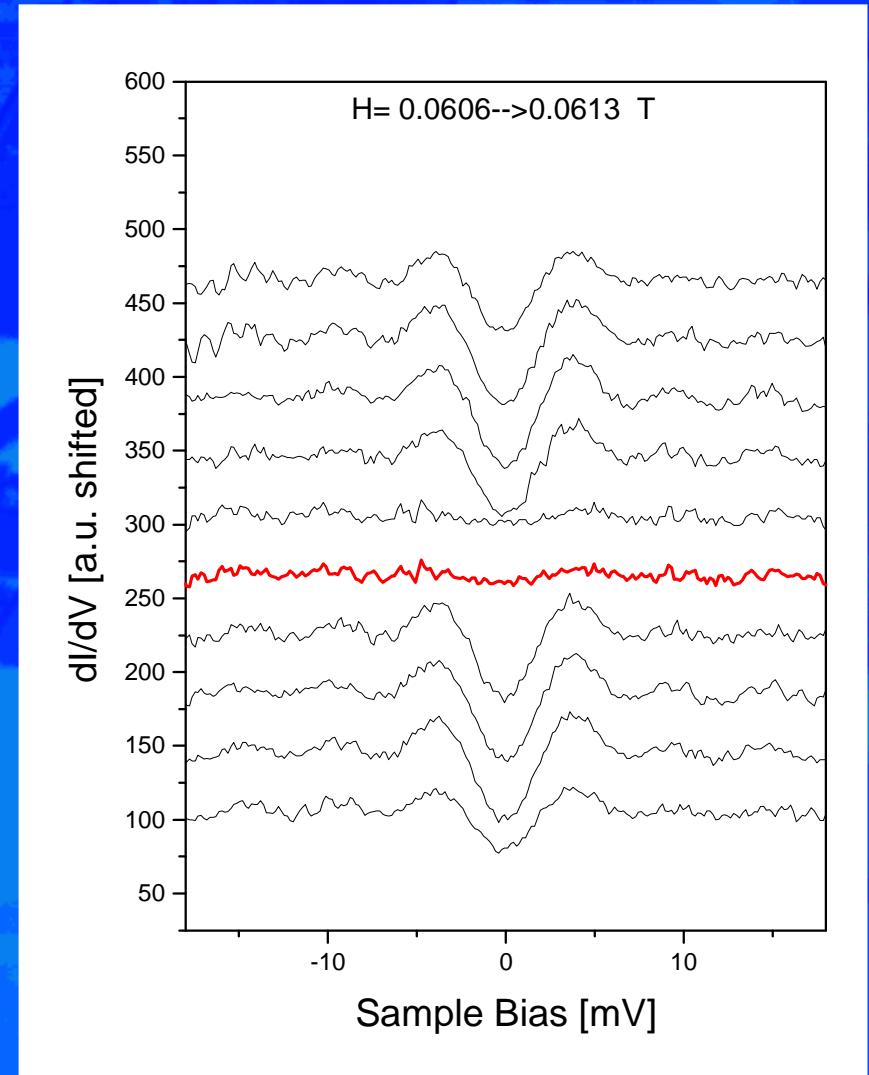
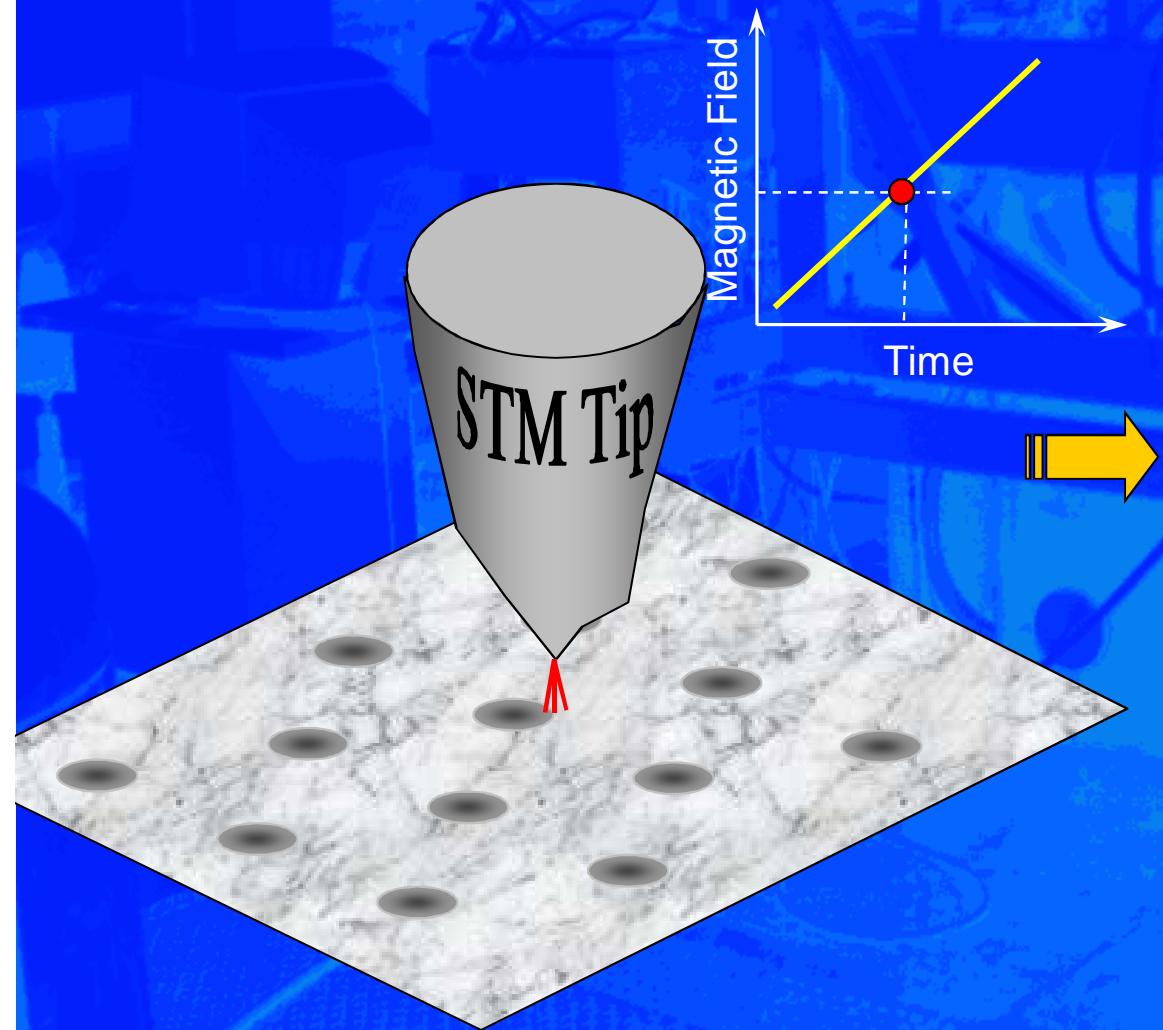
Lazy Fisherman : « Novel » Method of Vortex Studying

Local Spectroscopy



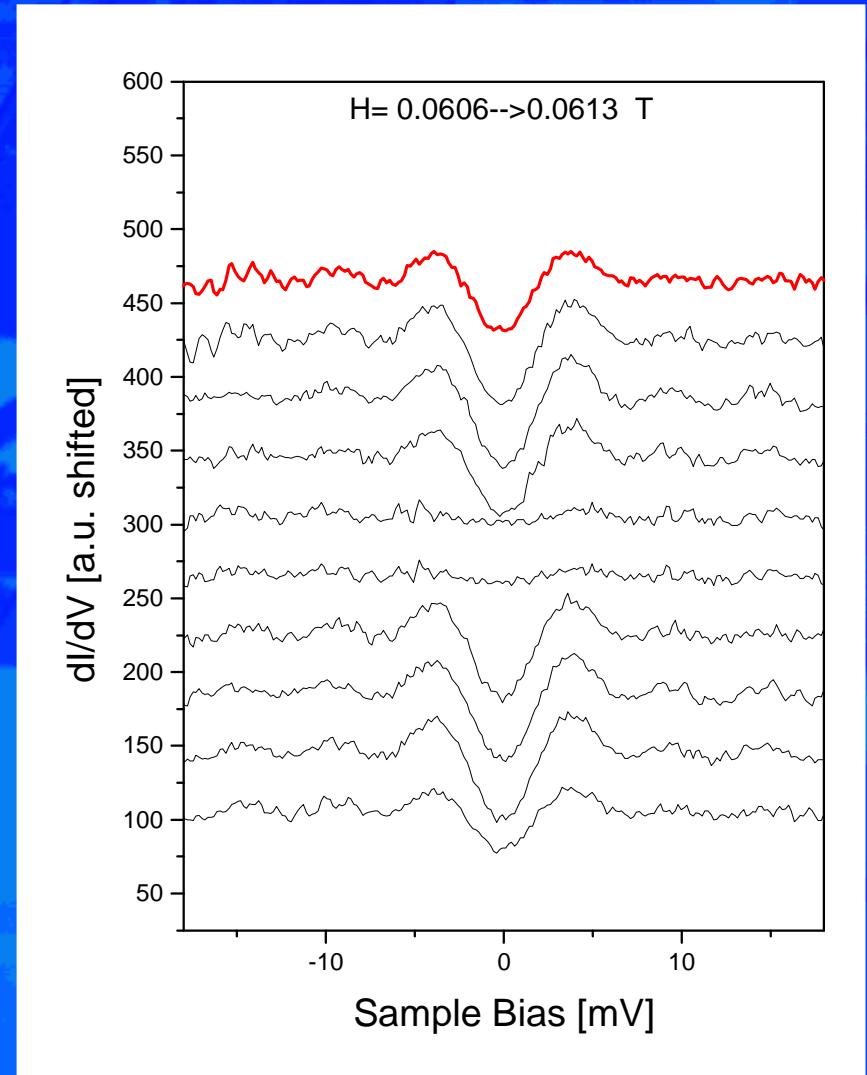
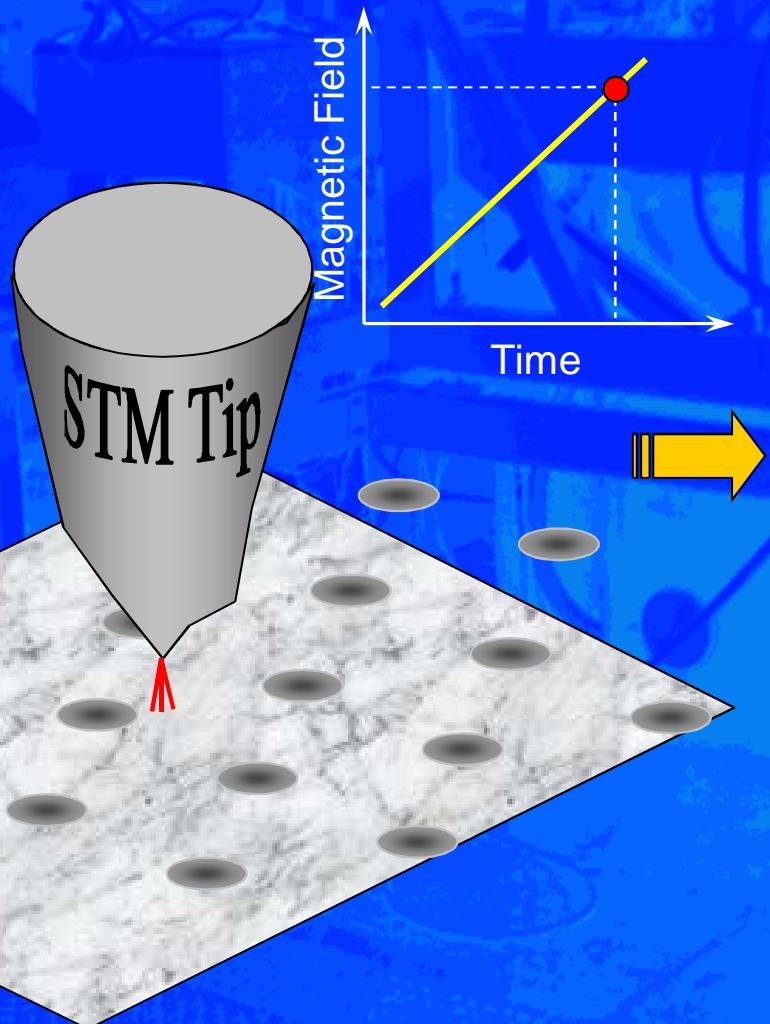
Lazy Fisherman : « Novel » Method of Vortex Studying

Local Spectroscopy



Lazy Fisherman : « Novel » Method of Vortex Studying

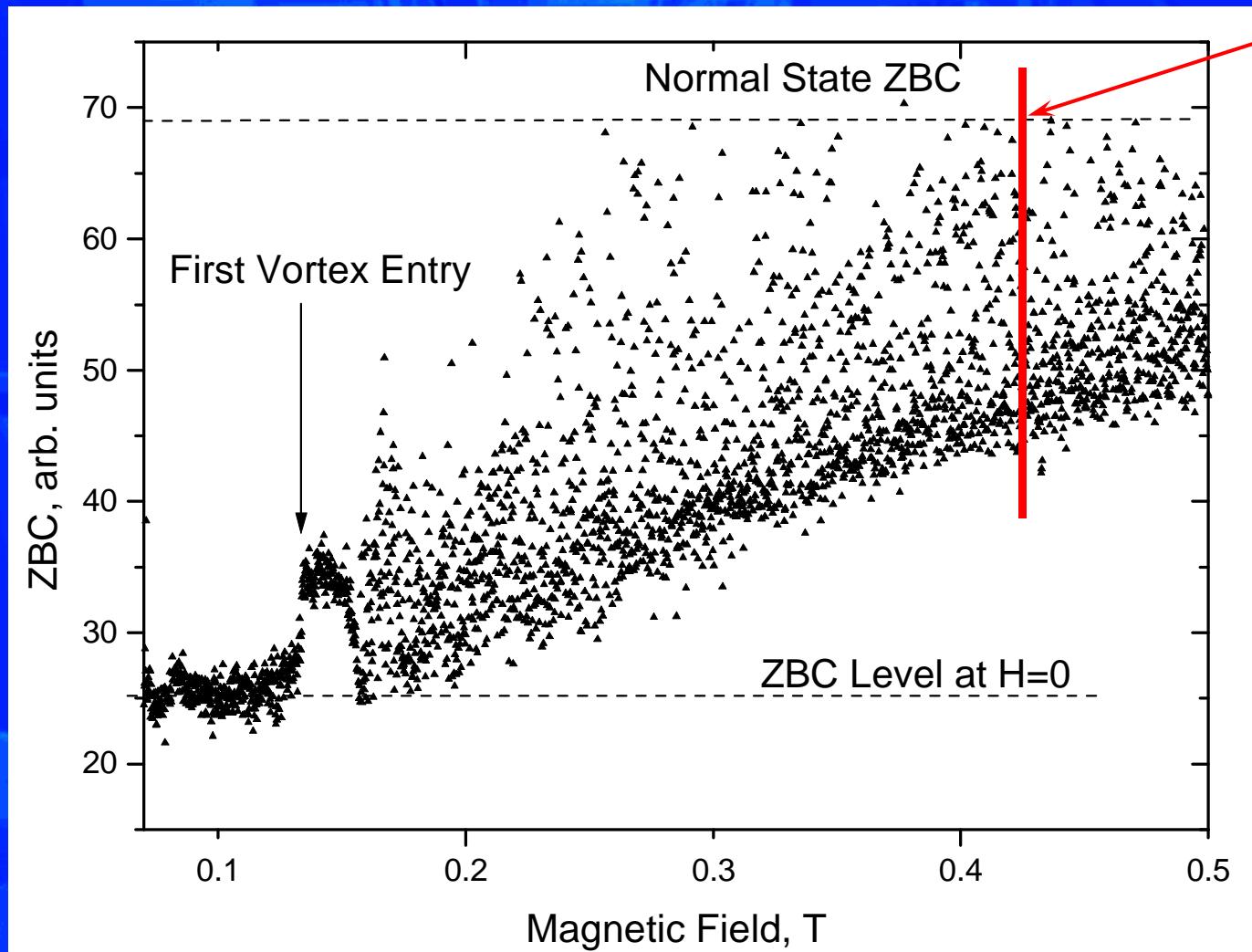
Local Spectroscopy



Lazy Fisherman : « Novel » Method of Vortex Studying

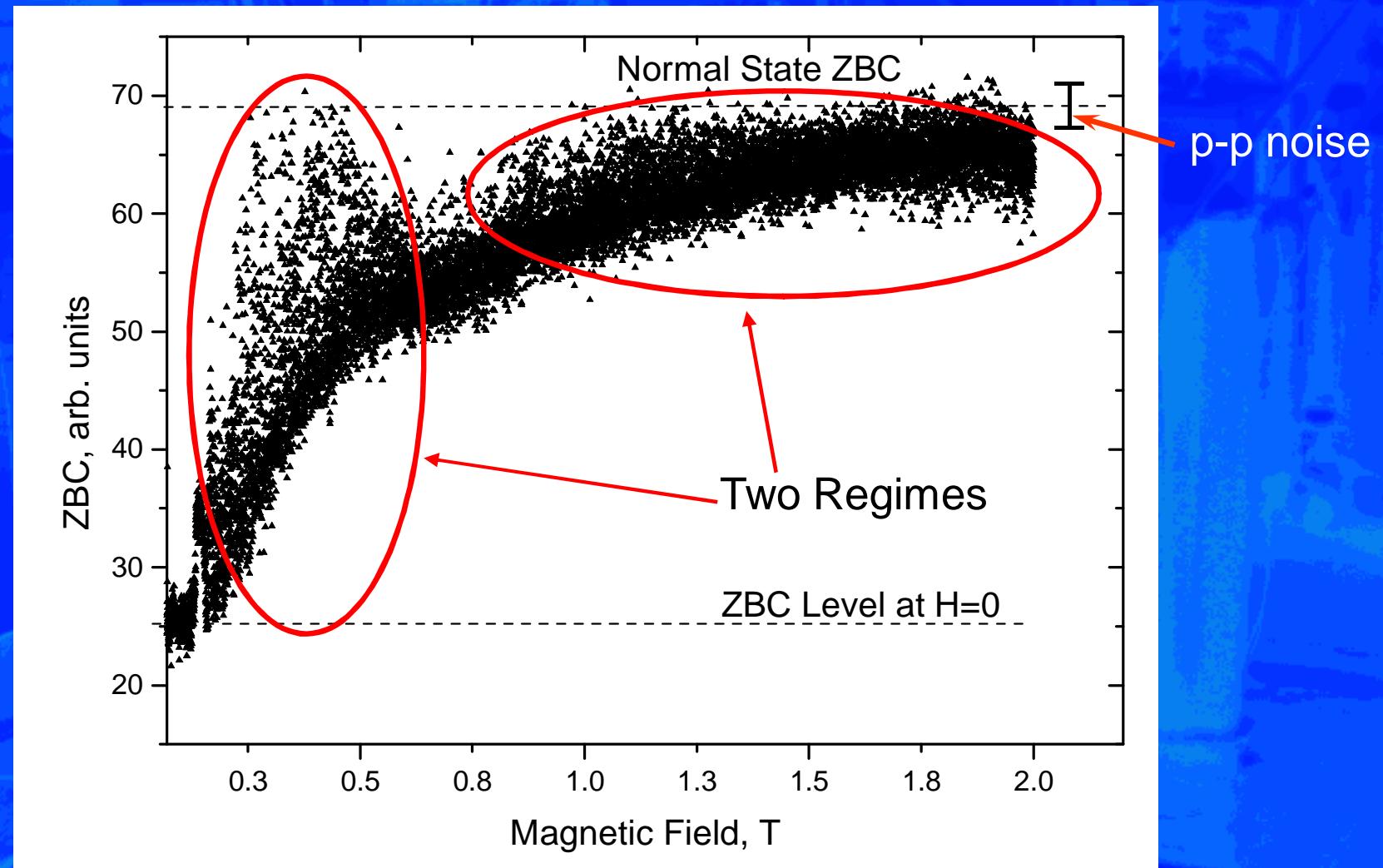
- Application to MgB₂

Profile Analysis : Size of the Vortex Cores



Lazy Fisherman : « Novel » Method of Vortex Studying

- Application to MgB₂

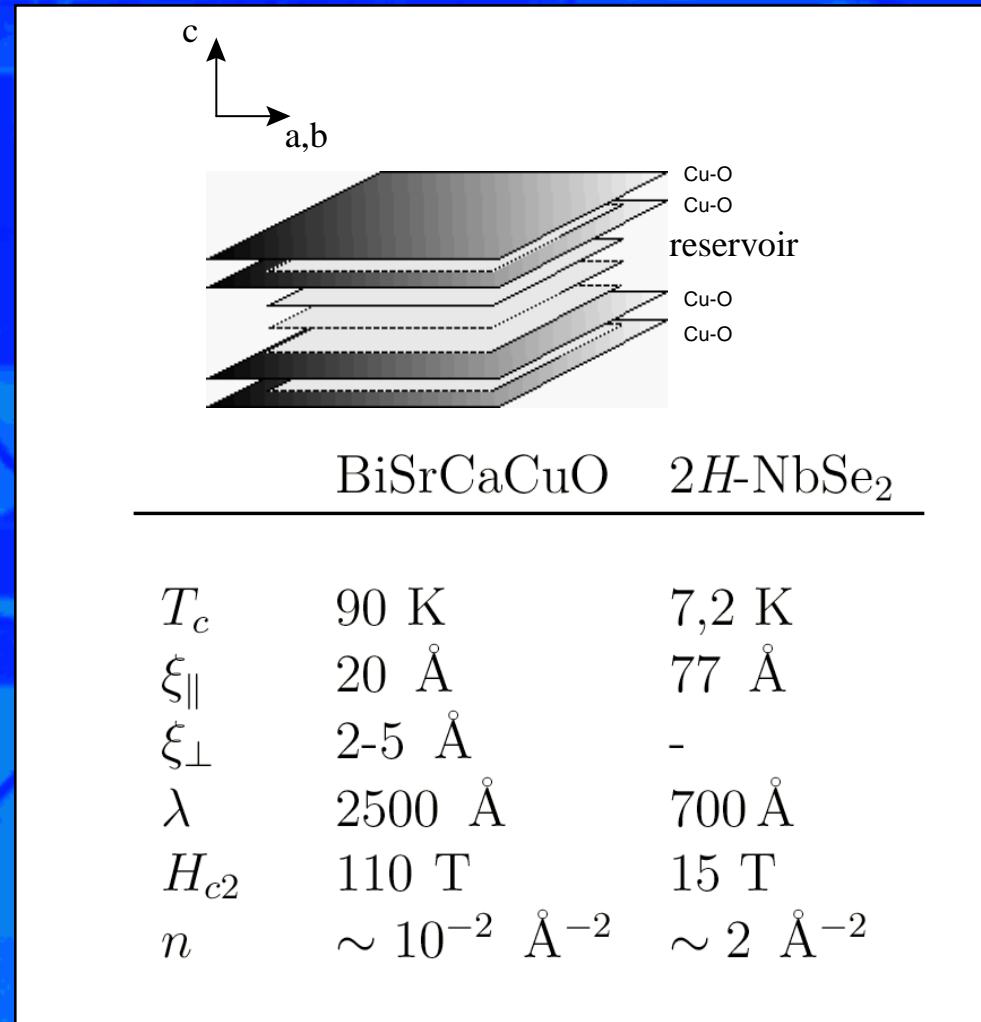
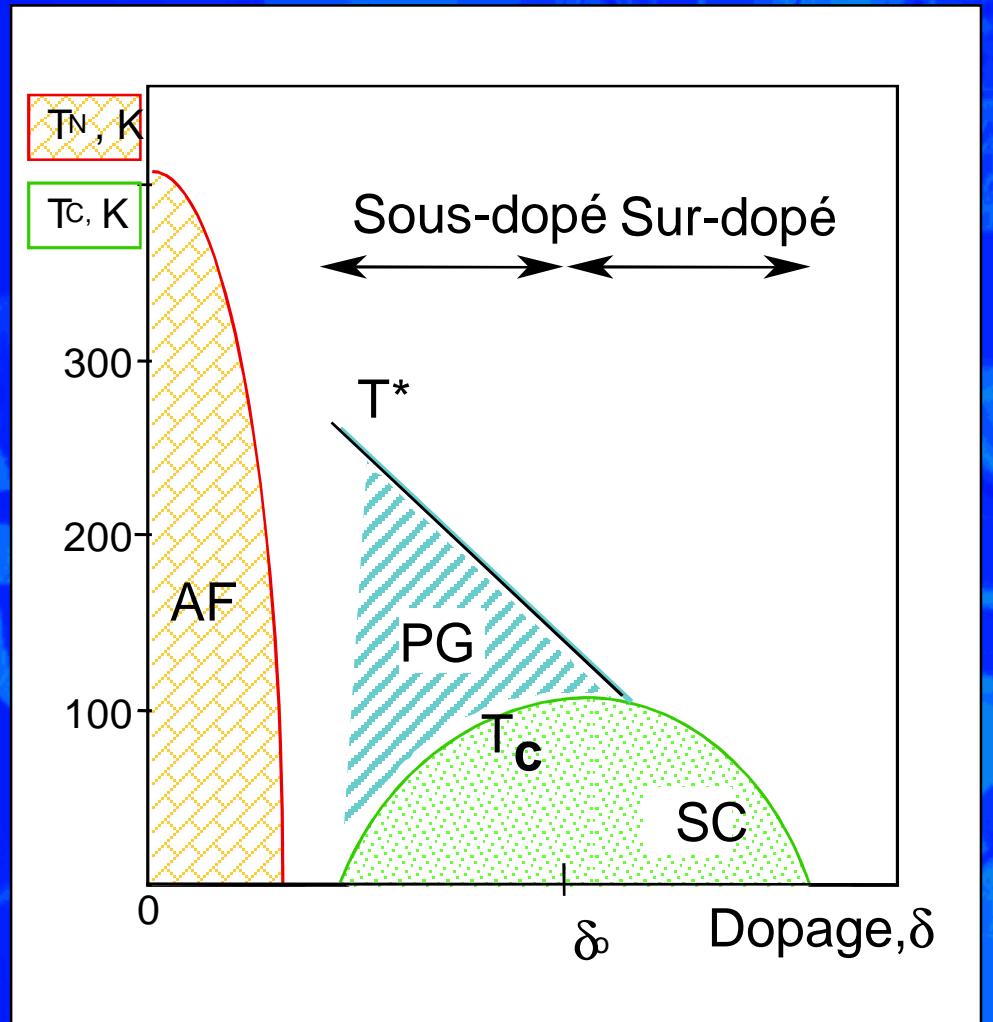


OUTLINE

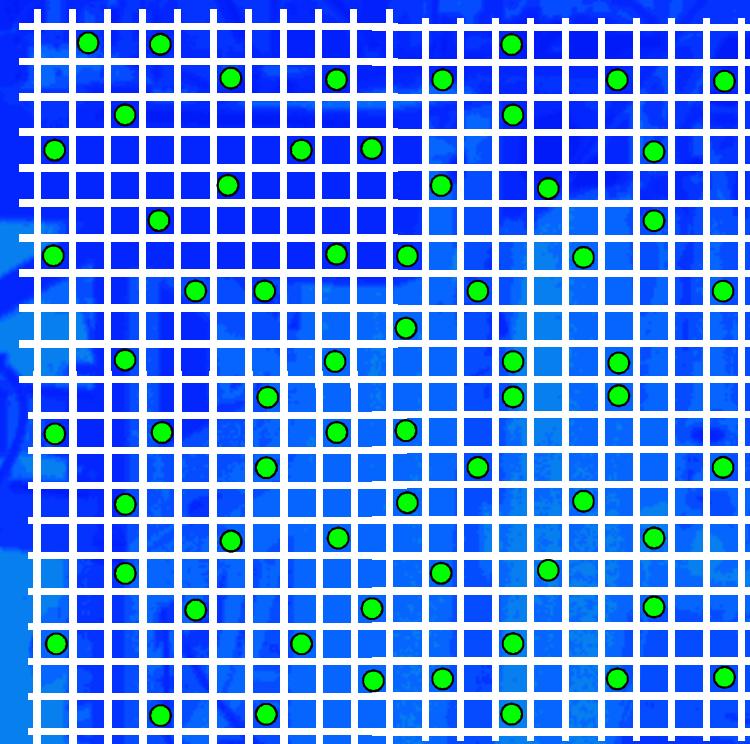
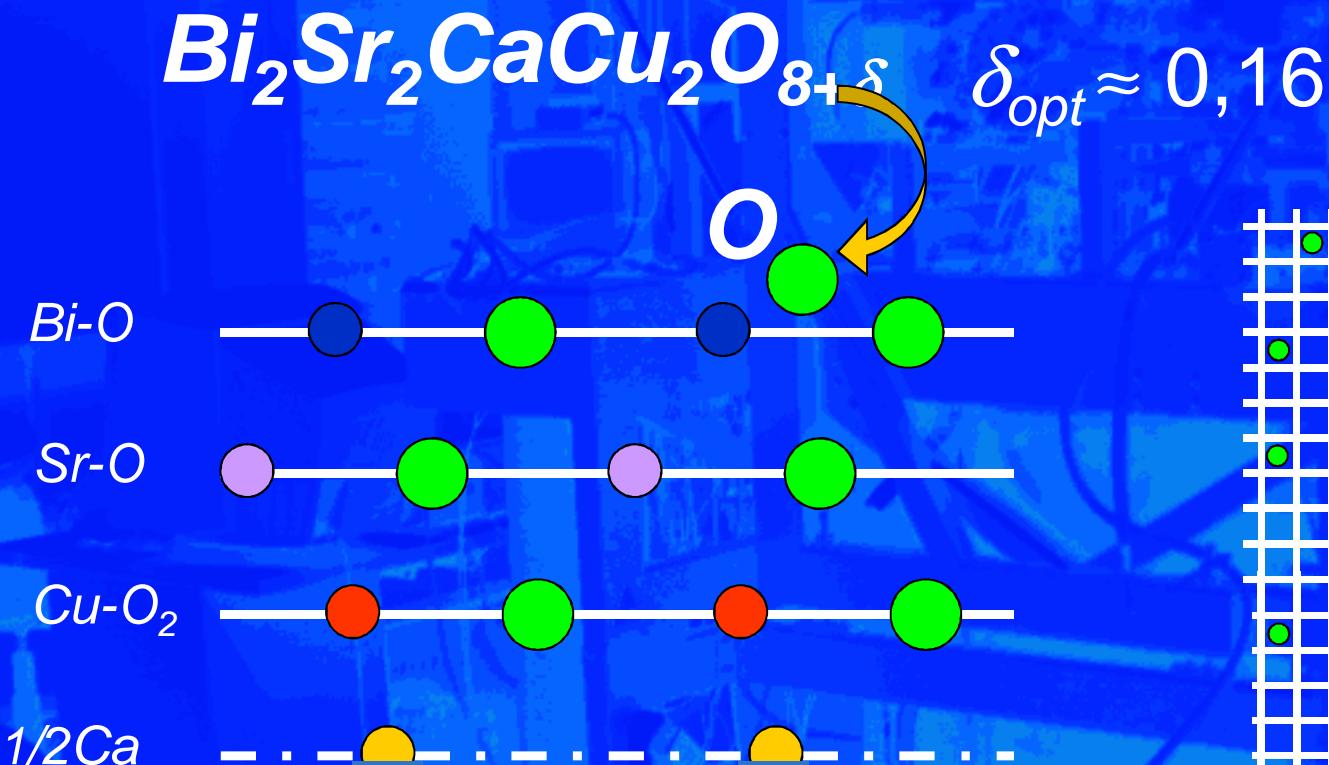
Scanning Tunneling Microscopy (STM) and Spectroscopy (STS)

1. Physical background
2. How to make it, how it looks like
3. Spectroscopy of Superconductors → HTSC
4. Other strongly correlated materials

Supraconductivité à haute Tc

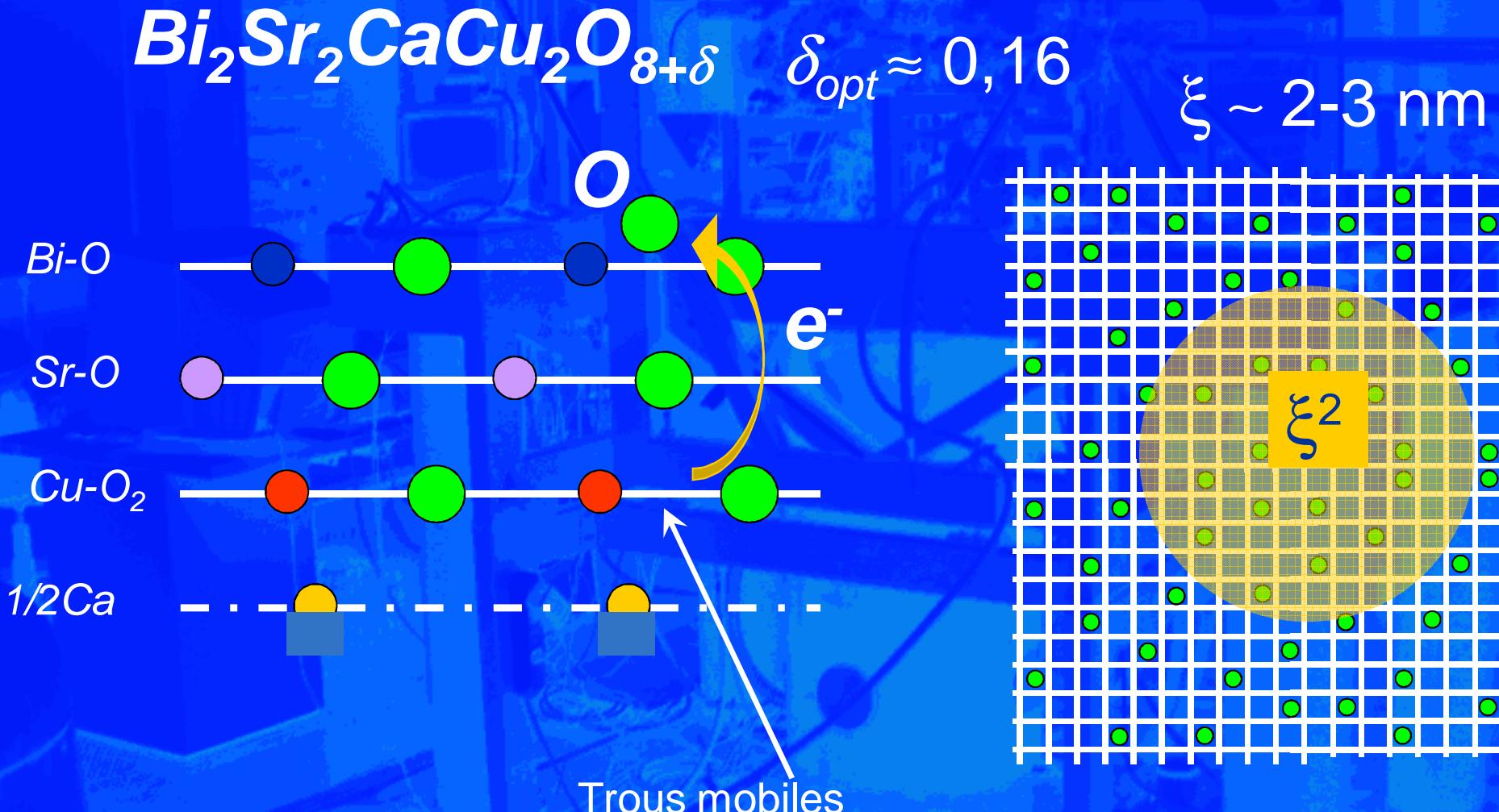


Supraconductivité à haute Tc



« OXYDES A PROPRIETES REMARQUABLES »
Aussois, 9-15 juin 2002

Supraconductivité à haute Tc



« OXYDES A PROPRIETES REMARQUABLES »
Aussois, 9-15 juin 2002

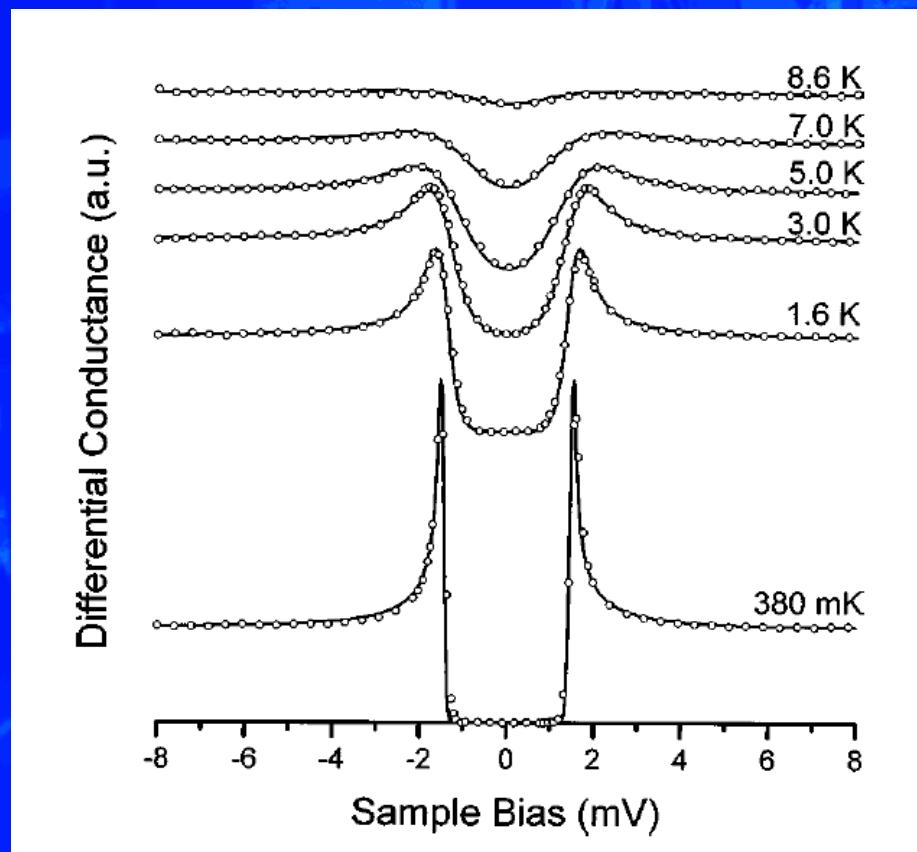
Supraconductivité à haute Tc : Gap



Jonction Nb-Au.

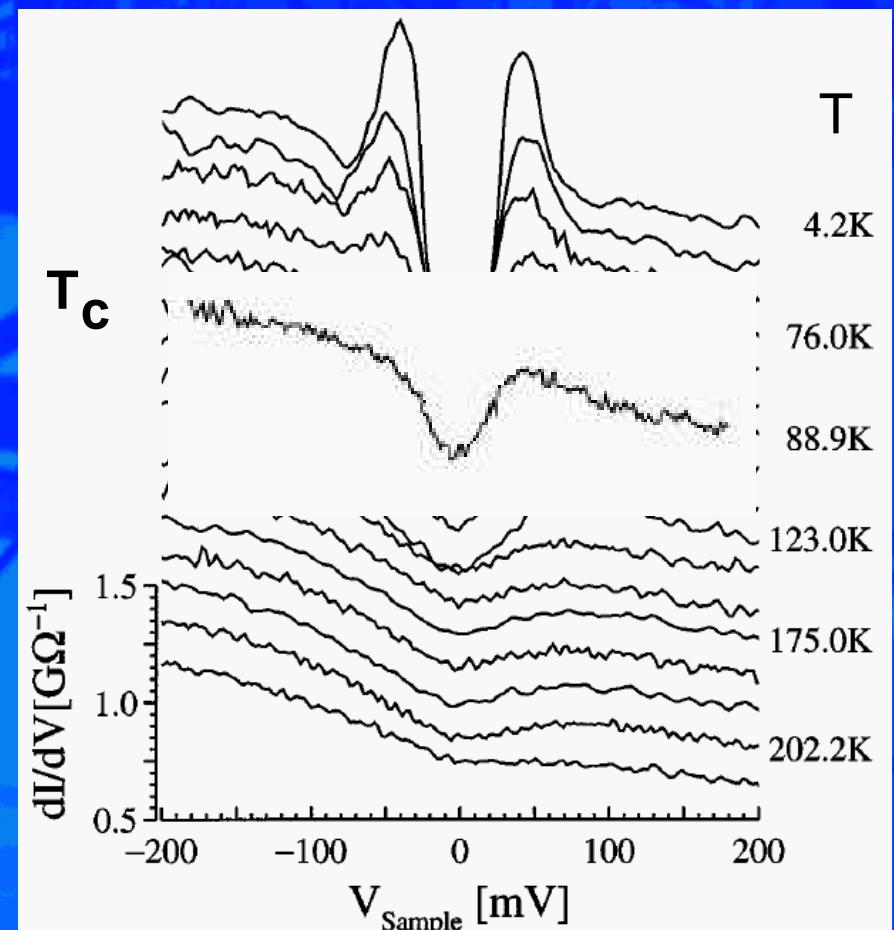


$Bi_2Sr_2CaCu_2O_{8+\delta}$ -Pt/Ir.



Pan, Davis,
Univ. de Berkley.(1998)

$$\frac{2\Delta(0)}{kT_c} \approx 3,5$$

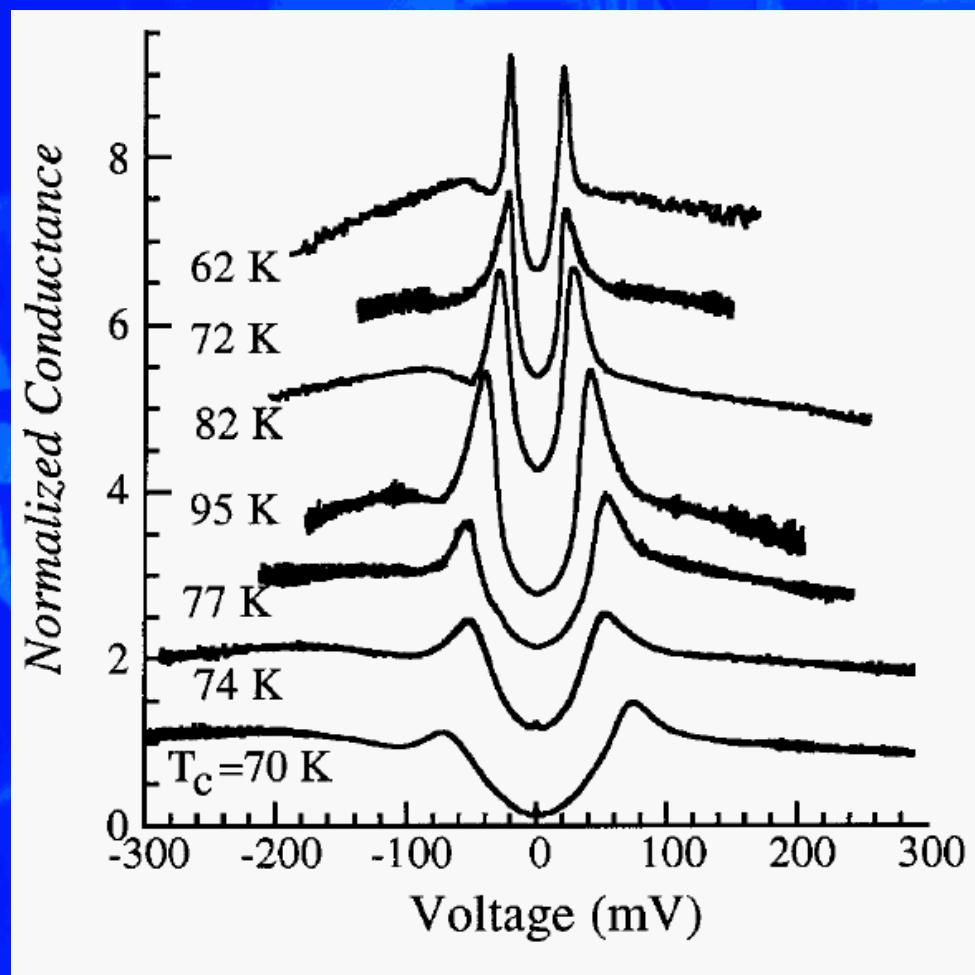


$$\frac{2\Delta(0)}{kT_c} \approx 38$$

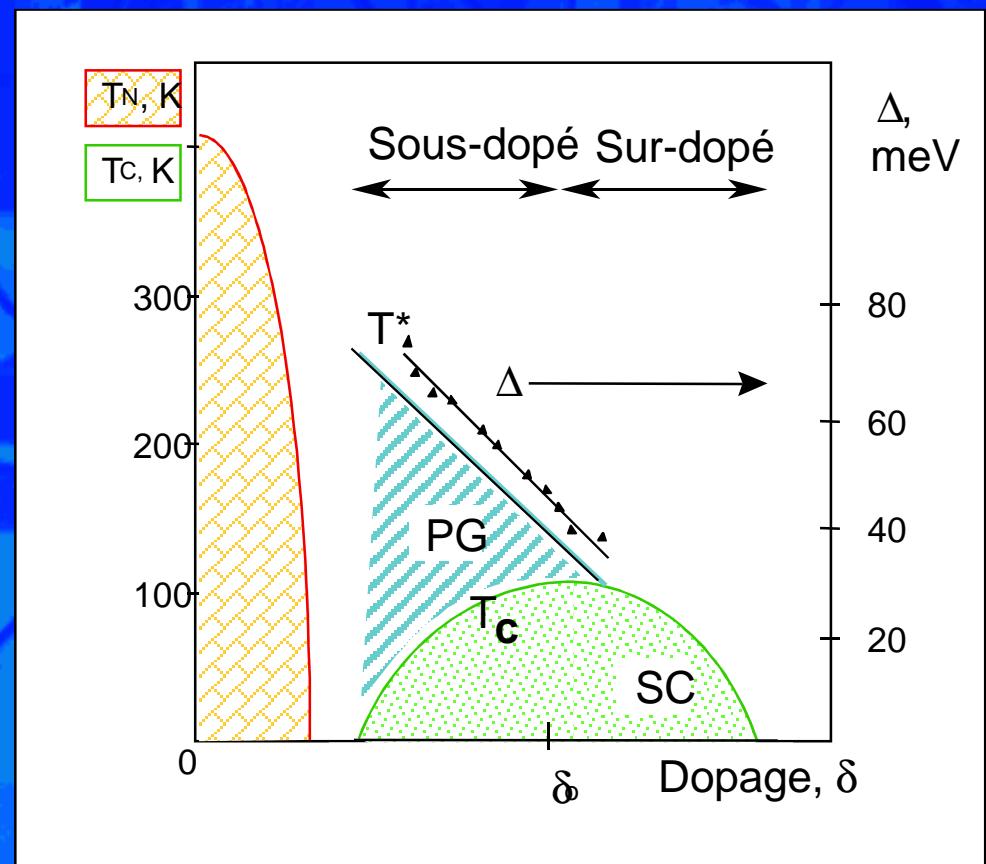
Ch. Renner, Ø. Fisher,
Univ. de Genève. PRL (1998)
Ecole MICO 2010

Supraconductivité à haute Tc : Dopage

● $Bi_2Sr_2CaCu_2O_{8+\delta}$ -Pt/Ir.



● Gap vs dopage.



Miyakawa, Zasadzinski,
PRL (1999)

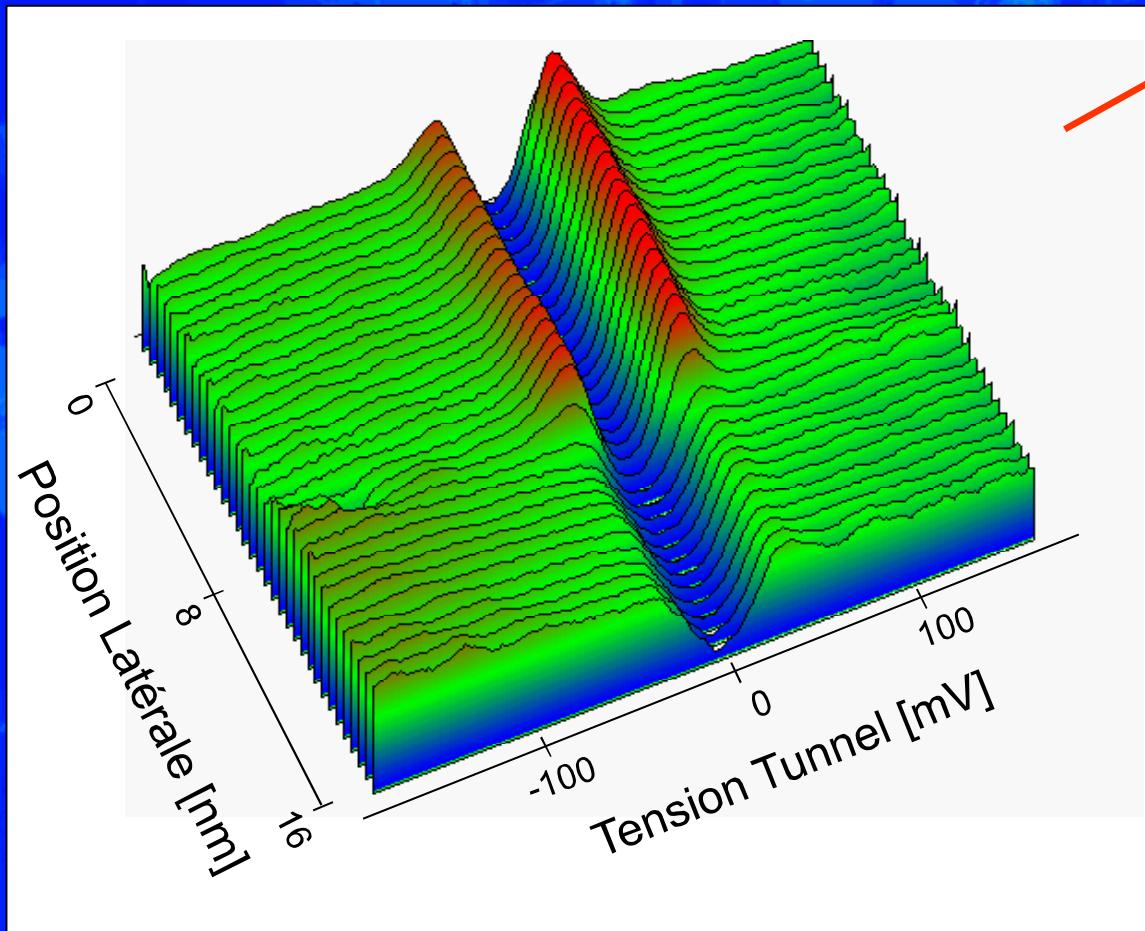
$$\frac{2\Delta(0)}{kT_c} \approx 5-60$$

!

Ecole MICO 2010

Pseudo-gap existe à basse température !

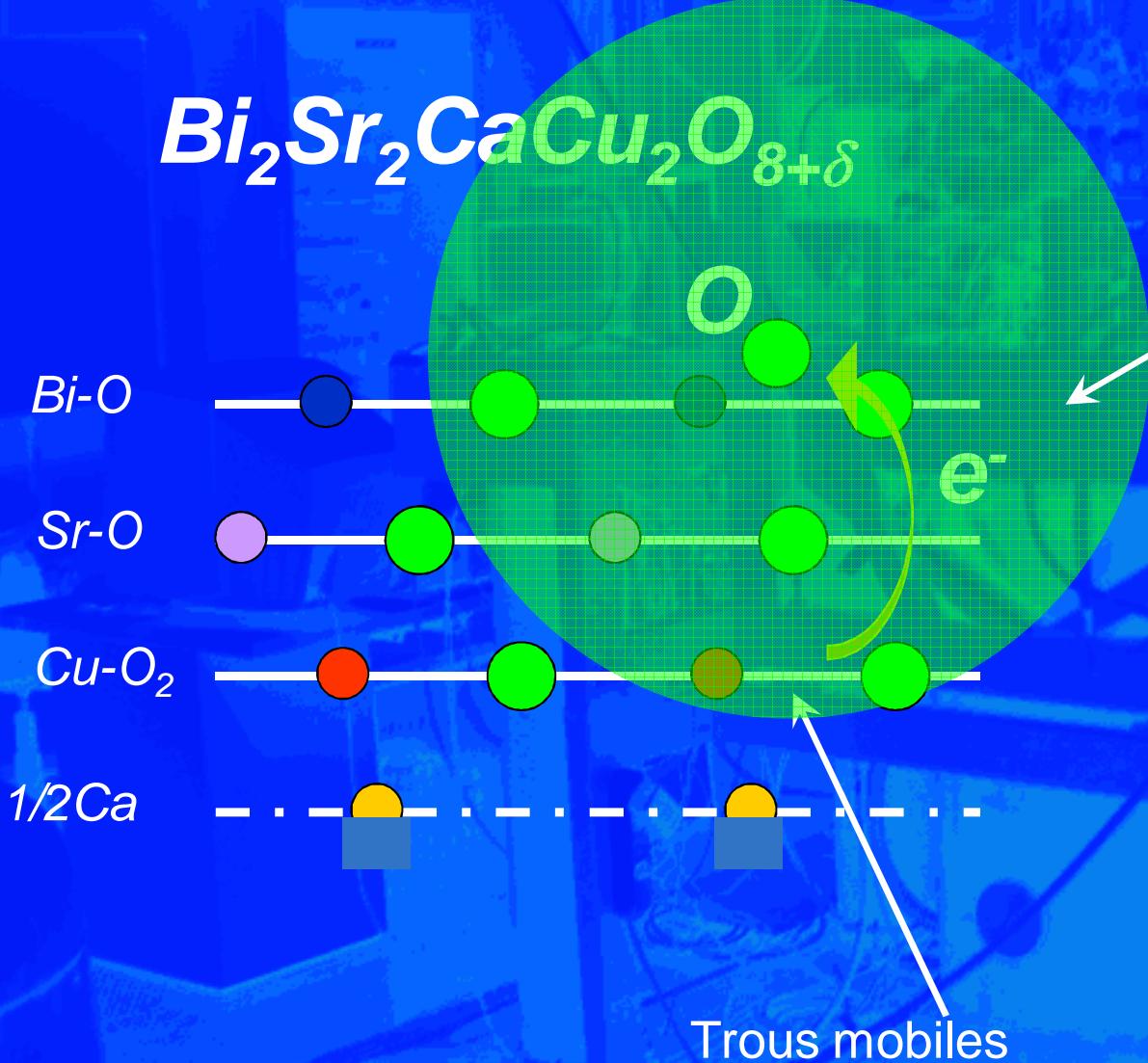
- Spectroscopie locale à l'échelle du nanomètre
(couches minces de $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$)



PG induit par le
désordre ?

Collaboration
INSP-ESPCI
et
INSP-CRISMAT

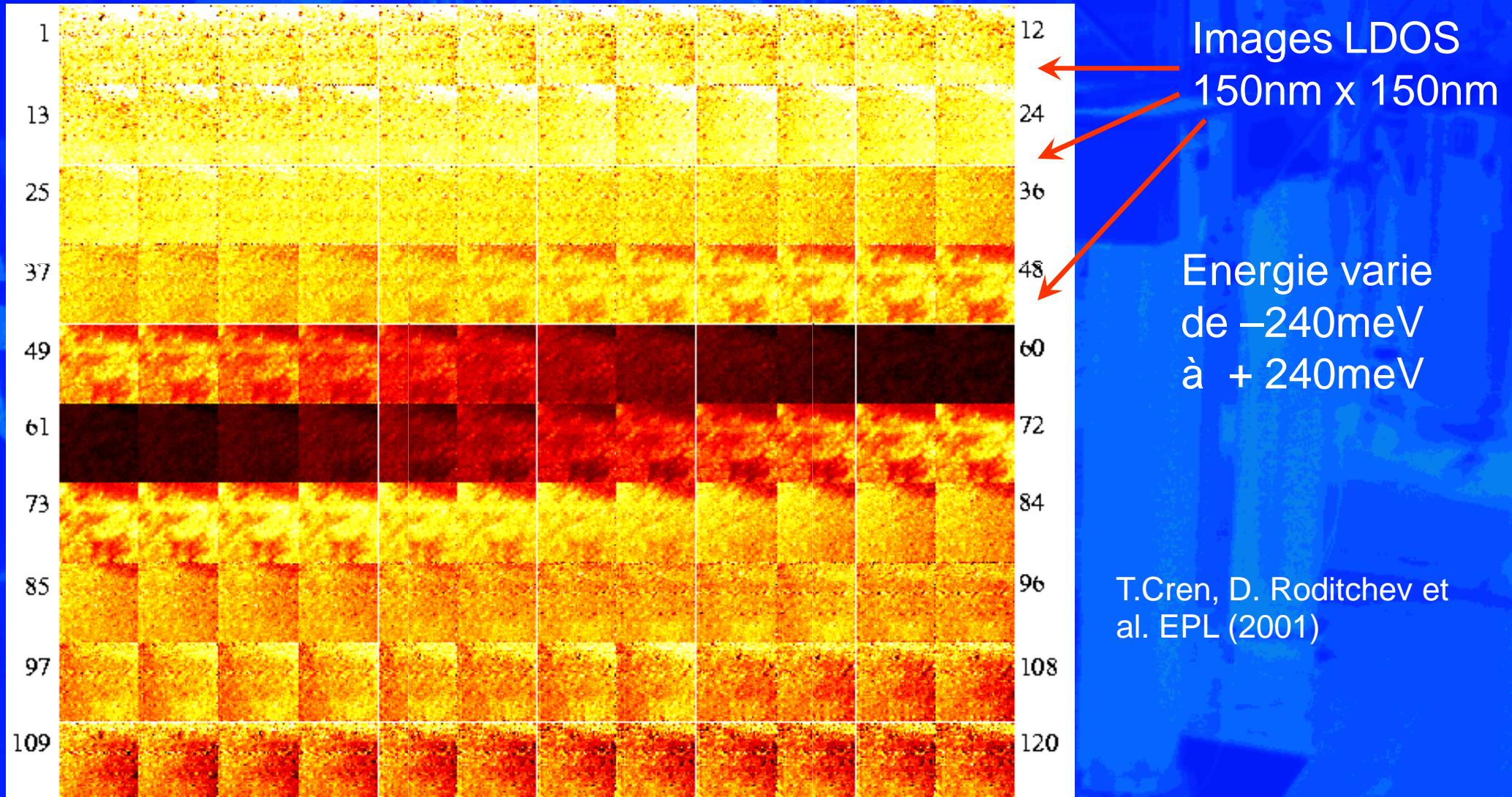
Cuprates et désordre



« OXYDES A PROPRIETES REMARQUABLES »
Aussois, 9-15 juin 2002

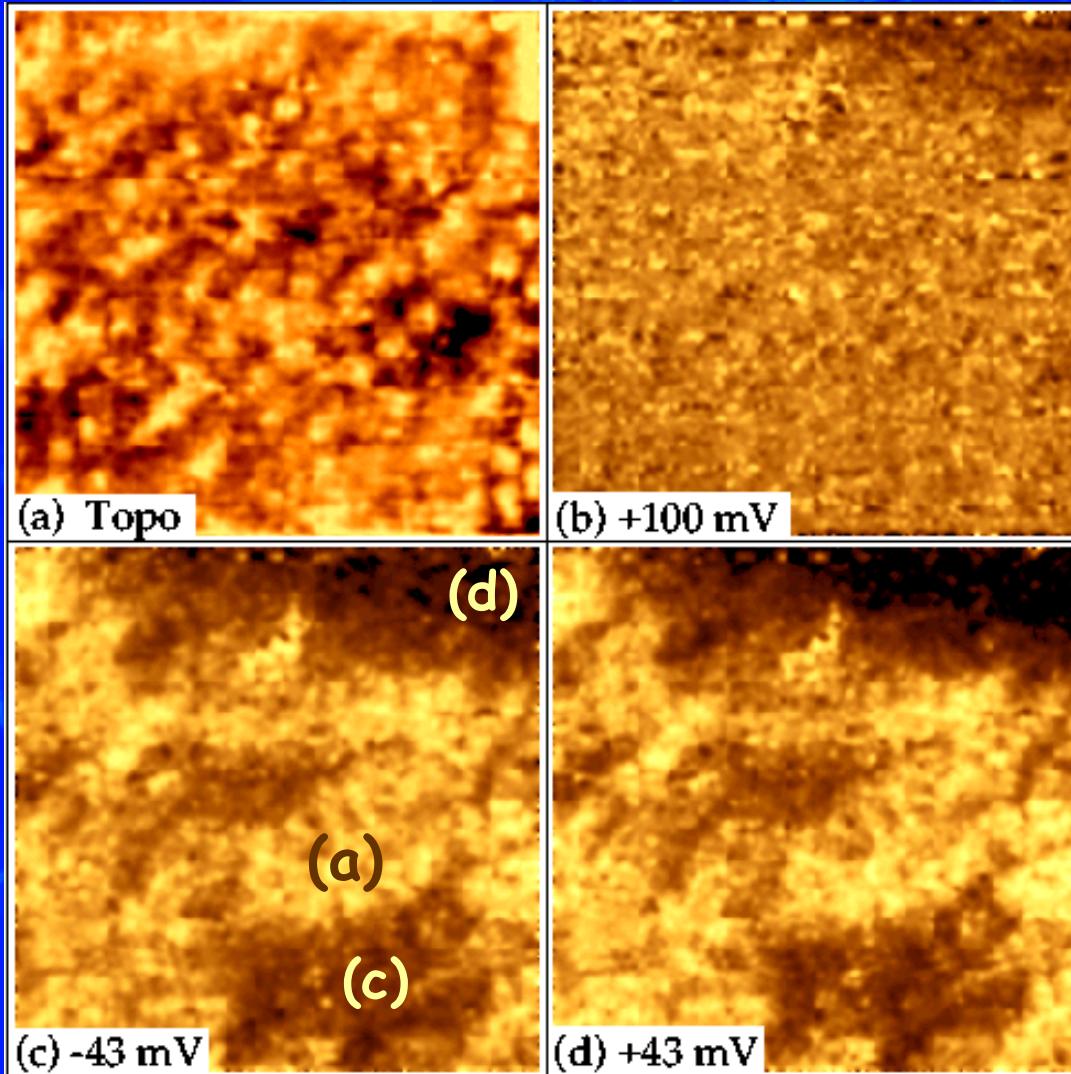
Influence du désordre substitutionnel

- STS sur des monocristaux de $\text{Bi}_{1-x}\text{Pb}_x\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$

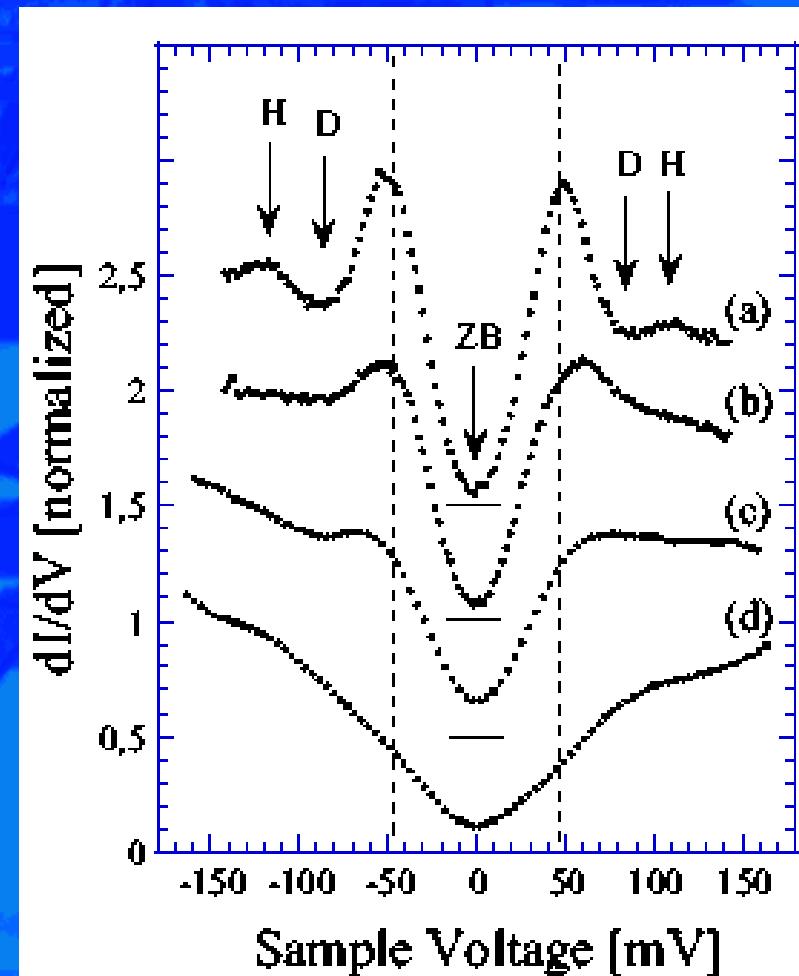


Influence du désordre substitutionnel

● Images LDOS 150nm x 150nm

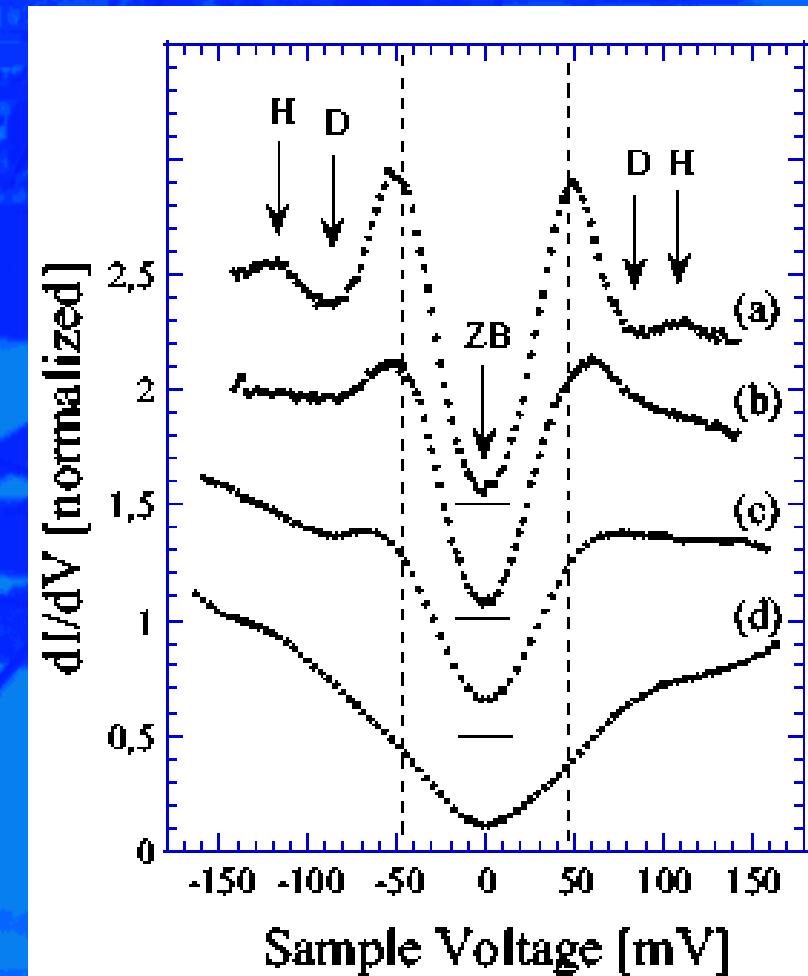
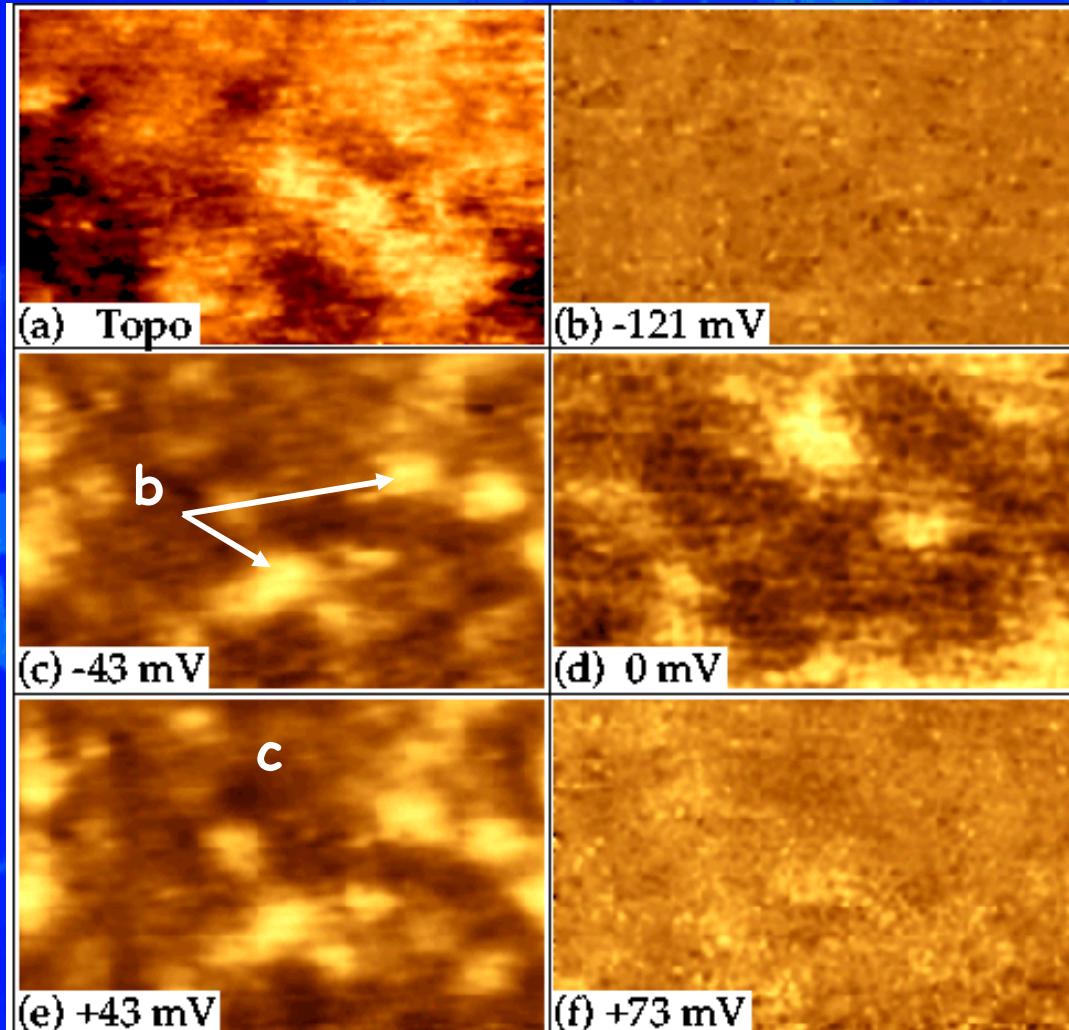


● Spectres tunnels



T.Cren, D. Roditchev, W. Sacks, J.
Klein, et al. Europhys. Lett. (2001)

Influence du désordre sur la supraconductivité des cuprates

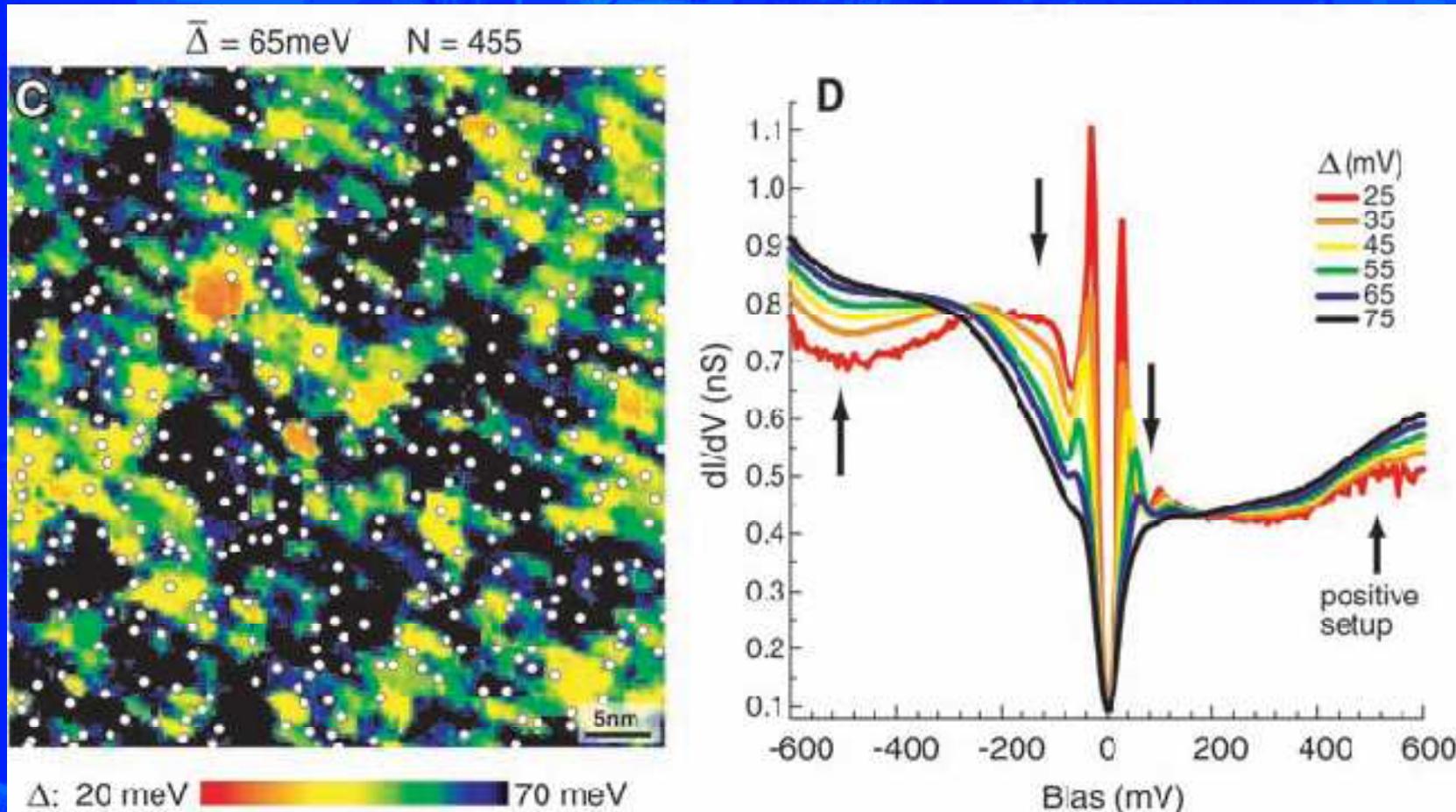


T. Cren, D. Roditchev, et al.
Europhys. Lett., 54(1), 84-90 (2001)

Collaboration ESPCI

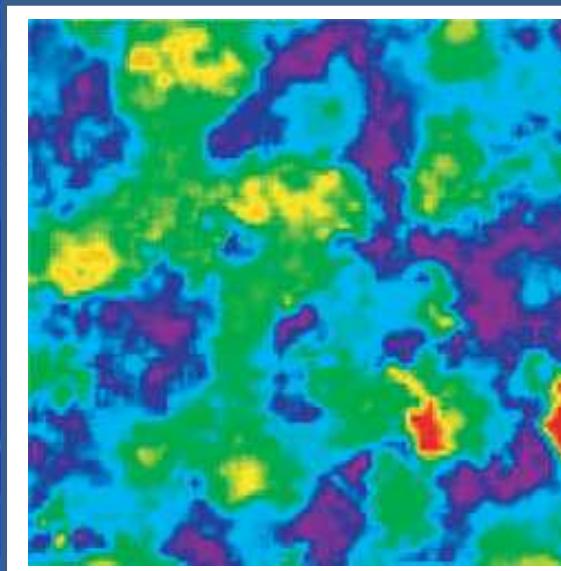
Ecole MICO 2010

Influence du désordre: 10 ans d'études en STM/STS

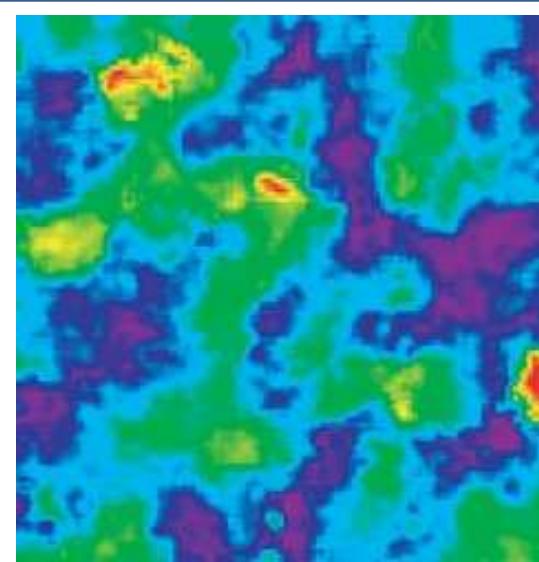


Cross-correlations among the simultaneously measured $Z(r, V)$, $\Delta(r)$ and $O(r)$ images showing a clear correlation between the gap, the oxygen and the structural disorders.

Influence du désordre: 10 ans d'études en STM/STS



Gap-map @5K, $T < T_c$



Gap-map @17K, $T > T_c$

Hudson et al., Nat. Phys. 3, 805 (2007)

Conclusion

STM/STS: Advantages:

Extraordinary spatial and energy resolution

Quasi-direct access to the DOS and its spatial extend

Possibility to explore (H,T) phase diagram

STM/STS: Weak Points:

Extreme sensitivity to the surface (states, quality etc.)

Poorly controlled \mathbf{k} -selectivity

High electric field may be perturbative

Experimentally heavy approach